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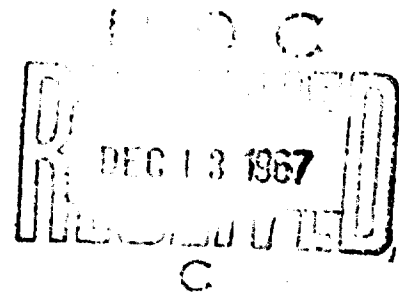
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EXPLOSION PHENOMENA INTERMEDIATE BETWEEN DEFLAGRATION AND DETONATION

- 1967 -

SCIENTIFIC AND TECHNICAL APPLICATIONS FORECAST



OFFICE OF THE CHIEF OF RESEARCH AND DEVELOPMENT
DEPARTMENT OF THE ARMY
WASHINGTON, D.C. 20310

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**EXPLOSION PHENOMENA
INTERMEDIATE BETWEEN DEFLAGRATION AND DETONATION**

**An
Army Research Office
Scientific and Technical Applications Forecast**

by

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Morton Collins**

**Esso Research and Engineering Company
Linden, New Jersey**

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FOREWORD

The documents known as Scientific and Technical Applications Forecasts (STAF) form a series designed to supplement the annual U.S. Army Long Range Technological Forecast. A STAF is intended to provide an insight into one specific field, and is for use by persons in and outside of the Department of the Army who have need for such background information. A STAF is primarily an encyclopedic summary of the current knowledge and a projection of the expected technological environment during the next 20 years. Its purpose is to allow scientifically, technically and operationally oriented individuals to communicate relevant ideas and learn of potentialities in the cited field.

STAF's, while generally comprehensive, are not exhaustive. Hence, the treatment may be properly considered an overall introduction to the current state of the art and an extrapolation to forecast the technological environment of 10-20 years in the future. An extensive bibliography is included in each STAF to document the current knowledge and to provide references for further detailed study.

This particular STAF on Explosion Phenomena Intermediate Between Deflagration and Detonation differs from most in that its primary emphasis is on theoretical interpretations rather than on technology. The technology of intermediate explosives was found to be virtually non-existent; consequently, the STAF analyzes the state of the art of intermediate phenomena in the context of a comprehensive review of modern reaction wave theory, and attempts to assess potential technology. An applications forecast and a research plan are presented, but they are necessarily more tenuous than ones for more highly developed fields. In view of the heavy emphasis on modern reaction wave theories and their problems, this STAF is primarily recommended to planners giving consideration to new areas for research and to personnel actively engaged in propellant and explosive research.

The conclusions of the authors are subject to modification in the light of new developments and information. Accordingly, readers are urged to submit comments in order to fill in possible gaps, report additional findings or applications, and suggest changes.

U.S. Army RDT&E agencies are encouraged to publish STAF's on specific scientific and technical subjects falling within their area of assigned responsibilities. Additionally, recommendations concerning desired subjects for STAF treatment are solicited and should be addressed to:

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SUMMARY

Intermediate explosives - defined as explosives whose propagation rates are faster than those of conventional deflagrations and slower than those of conventional detonations - would presumably produce pressures and reaction times also intermediate between those of deflagrations and detonations. Such materials could have numerous military and industrial uses because they would enable simplification of mechanical arrangements in certain existing systems and would offer some new capabilities.

No true intermediate explosives have been developed and proven as of this writing (1967); but a number of examples of apparently intermediate velocity and pressure phenomena have been observed, and at least some of them show promise of being developable into practical intermediate explosives. One example, consisting of ammonium nitrate granules suitably coated with, for example, nitroglycerin, is actually a detonating explosive; but the detonation propagates at approximately 1/6 the velocity of most detonations. The inventor is Dr. M. A. Cook of Intermountain Research and Engineering Company, Salt Lake City, Utah. Another example consists of a proprietary (and unrevealed) monopropellant mixed with stoichiometric amounts of inorganic oxidizer. This composition is reported by the discoverer, McCormick-Selph Company of Hollister, California, to propagate at controllable rates up to 280 m/sec., which is much faster than any previously-known, stable deflagration, but an order of magnitude slower than normal detonations. Most of the other intermediate-type phenomena which have been observed appear to be non-ideal detonations which are either unstable or require tricky geometrical configurations which make their ultimate practical utility problematical.

Intermediate explosives can only be understood in the context of the state of the art of explosives in general; consequently this report presents a review of reaction wave theory brought up to date (1967), followed by a description and interpretation of apparently intermediate phenomena which have been observed. The review of reaction wave theory, while believed to be comprehensive, does not attempt to be exhaustive. It attempts to give the reader an appreciation of the field as a setting in which to consider intermediate explosives. A very extensive bibliography, both cited and general, is given for investigators wishing more detail.

Reaction wave theory does not clearly account for intermediate phenomena. Current theory predicts two and only two stable combustion regimes, deflagration and detonation, with any and all intermediate regimes being unstable or, at best, metastable. However, the theory is far from complete, most of the boundary assumptions are acknowledged oversimplifications, and many of the models are only qualitative. The real world is much more complex, and the current theories are adequate only in a limited number of circumstances - not including those of major interest here. This does not imply that the current theories are incorrect; only that a more comprehensive theory must be developed, of which the current theory will be a special case. It should be realized that intermediate velocity reaction waves are in reality a part of the continuum of reaction waves which encompass all reactive flow. Intermediate phenomena are empirical facts; and rather than being explained by current theory, they will help to refine the theory, particularly the sometimes tacit assumptions implicit in the models.

This review classifies the observed, apparently intermediate, phenomena into four categories:

- Ideal detonation on low density media
- Nonideal detonation
- LVD in liquids and solids
- Convective deflagration

and discusses both the phenomena and their interpretations within this framework.

A research plan to develop intermediate explosives has two aspects: (1) research to understand the phenomena, and (2) R&D to develop useful systems. The former is a basic program aimed at elucidating the phenomena which have been observed and the mechanisms by which they operate. Hopefully, understanding will lead to application; but understanding itself is the goal. The second is an applied program to take the most promising examples already observed and evaluate their practical utility by large-scale, and more intensive, testing.

The incentive for carrying out the research plan is essentially that of keeping the technology ahead of the requirement. Most of the applications engineers consulted felt that they could meet all of their current requirements with current explosives. This is somewhat too limited a view - there are current systems which would be simplified or improved by intermediate explosives - but it is nevertheless true that a really pressing need for intermediate explosives is contingent upon requirements not yet issued.

The information and views in this STAF are based upon a year-long study of the explosives field. It included the major open literature of the world, the classified literature in DDC and in leading military laboratories,* visits to many of the leading researchers of this country and Europe, and interviews with a considerable sampling of applications people both in the military and in industry. Significantly, the most exciting leads were found on the periphery of the explosives field, rather than directly in it; and it is in peripheral fields that further new leads should be sought.

*This document does not reveal any classified information.

1. INTRODUCTION

Intermediate explosives - materials exhibiting stable reaction rates and pressures intermediate between those of deflagrations and detonations - constitute a new class of explosives which, if available, would have a number of uses in both military and industrial applications.

1.1. Definition of What an Intermediate Explosive Would Be

An explosive may be defined loosely as a material which reacts rapidly - often violently - to produce large quantities of hot gases in a confined space in a short time and capable of doing useful (including destructive) work. For present purposes, it is intended to exclude steam explosions, pressure vessel ruptures, etc., and to focus attention upon chemical explosives as such.

Deflagrations

Deflagrations are burning phenomena whose propagation rates are controlled by transport processes and by chemical kinetics. They are characterized by the dependence of the linear burning rate on the ambient pressure, and their reaction rates are low compared to those of detonation. In the condensed phase, propagation rates in void-free materials range from a fraction of a centimeter per second to about 12 centimeters per second at 1000 psi. Laminar burning in gases at one atmosphere ranges from 3 cm/sec for propane-air to about 75 cm/sec for hydrogen-air. In turbulent gases, the rates may reach hundreds of meters per second.

In the unconfined state, the pressures generated by deflagrations are low, on the order of inches of water. If the material is confined, as in a gun, the pressure is essentially that given by the gas law for the appropriate adiabatic heating and increase in the number of moles of gas.

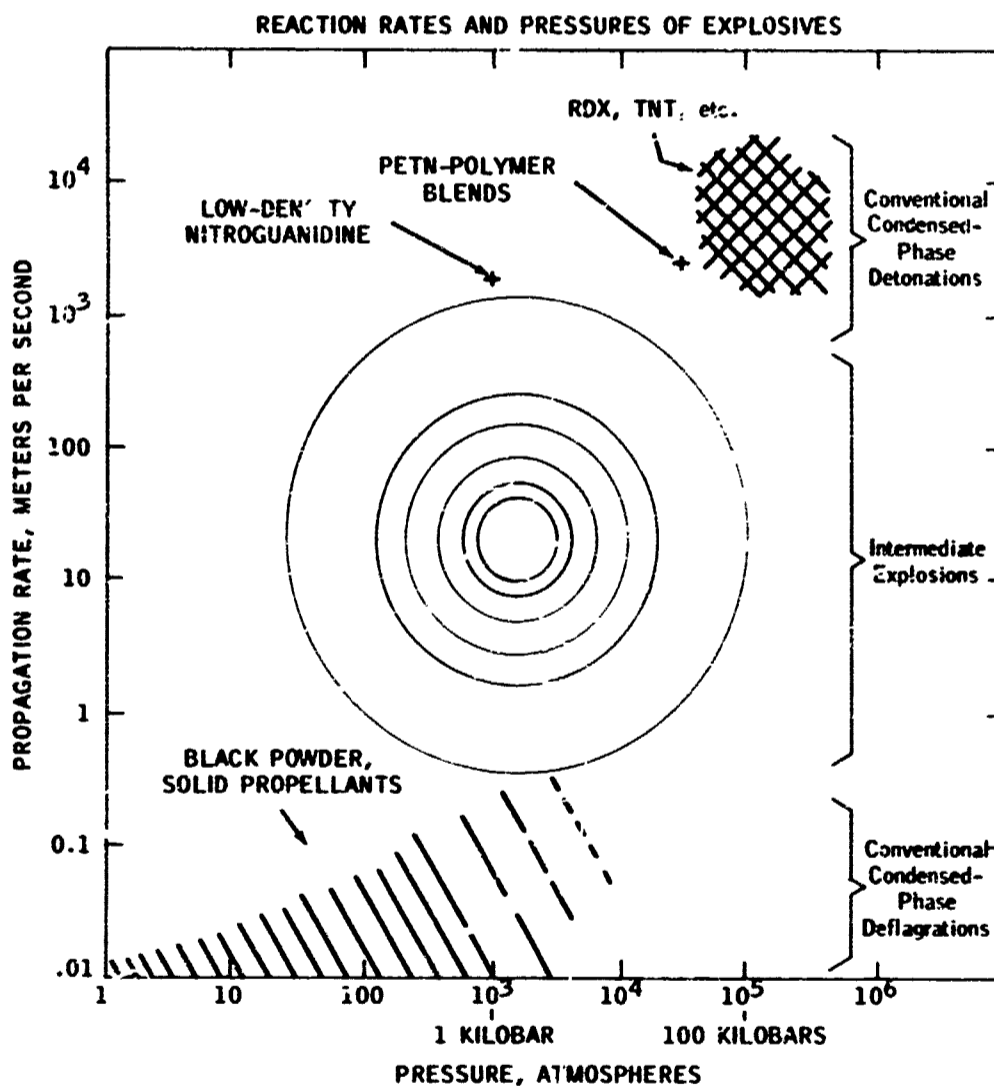
Detonations

Detonations are reactive wave phenomena whose propagation is controlled by shock waves. Theoretical analyses assume that reaction rates are essentially infinite and that chemical equilibrium is attained. Therefore, the actual propagation rate is considered to be governed solely by thermodynamics and hydrodynamics. The propagation rates of detonations are orders of magnitude higher than those of deflagration, i.e., thousands of meters per second.

The pressure generated in detonations is very high, whether confined or unconfined, due to the very high propagation rate. The gases have not had time for expansion; a pressure pulse can only move at sonic velocity, and the detonation front moves at supersonic velocity.

Intermediate Explosions

There is a gap of several orders of magnitude between the propagation rates of conventional deflagrating explosives such as black powder or double base propellants (cm per second) and conventional detonating explosives such as TNT or RDX (thousands of meters per second). There is a similar gap between their unconfined reaction pressures. These relationships are illustrated diagrammatically in the following figure, where reaction rates are plotted against reaction pressures. Deflagrations fall into the lower left-hand corner of the diagram, with low pressures, low rates and long action times. Detonations fall into the upper right with high pressures, high rates and short action times. Intermediate explosions would fall between, with pressures of a few tens of kilobars, propagation rates of a few hundreds of meters per second, and correspondingly intermediate action times.



Thermodynamic theory, based on a fixed slate of products, indicates that only two stable reaction regimes are possible - deflagration and a single high order detonation. In practice, however, intermediate rate reactions are frequently observed in condensed explosives. They are not well understood, and have not yet been applied in practical devices, primarily because they have not yet exhibited sufficient stability of properties. However, they are worthy of study, because they offer new capabilities not possessed by conventional low or high explosives.

1.2. Possible Applications

Intermediate explosives offer a variety of new capabilities, based on their slower rates and lower but sustained pressures as compared to detonations. A really detailed catalog of uses is not possible at this time; the following sketches* are intended to indicate the possibilities and to stimulate the thinking of applications engineers.

- New ranges of timing and delay fuses - Current fuses are either detonating lines with propagation rates of about 6000 m/sec. or deflagrating lines with propagation rates of a few cm/sec. This makes microsecond-range delays and second-range delays available, but millisecond delays need improvement. At least two commercial companies are working on this, one by manipulating environmental parameters such as column diameter, and the other by developing new explosive compositions.
- Non-brisant fuse trains - Conventional detonating fuse, such as is used to conduct an initiation from one part of an air or space craft to another, must be heavily armored in order to avoid damaging panels and structural members which it traverses. An intermediate explosive loaded fuse would have less incidental damage potential.
- Non-brisant explosive separation devices - Explosive bolts and explosive hatch jettison charges based on high explosives throw too much shrapnel and do too much damage beyond the area which is to be cut. A somewhat lower pressure exerted for a longer time would moderate the blast effect and permit more efficient coupling of energy.
- Explosively-actuated tools - Cable cutters, rivet setters and the like employ an explosive charge which drives a pneumatic or hydraulic transformer to convert a short, sharp push to a more prolonged one. An intrinsically intermediate explosive could eliminate the transforming linkage by delivering a gentle, prolonged impulse directly.
- Chaff ejectors and gas generators - In aerial countermeasures, there is a need to eject radar decoy chaff with a very short response time. Cartridges are sometimes too slow and detonating cartridges do too much incidental damage. Intermediate rates and pressures would be more favorable.
- Mining, where a prolonged heaving action is desirable - In the mining of coal or rock, high explosives tend to give too much shattering of the excavated material. Black powder and Cardox have been used for this reason, but have other disadvantages. The prolonged pressure pulse of an intermediate explosive would be of clear value.

*These thoughts are distilled from a great many interviews with many different people, and it is not possible to indicate or even to know all the initial sources. Some of the ideas are our own; and, in any case, the same ideas kept recurring in conversation after conversation. A list of individuals and organizations interviewed is appended.

- Metal forming and welding - This is very much an art, and elaborate stand-off charges and air and water moderators are used to tailor the explosive pulse to the stress-strain characteristics of the metal being formed. With an intermediate explosive, one could use contact charges and eliminate much of the mechanical complexity of current set-ups.
- Single-grain gun propellants - Current gun propellants, from cannon powder to shotgun loads, achieve their short action time by exposing an extremely high surface area. This is achieved by forming the "powder" into hundreds of thousands of tiny individual grains, usually cylindrical and often perforated. An intermediate explosive, that is, an extremely fast-burning propellant, could be cast or pressed as a single grain with attendant manufacturing economies. The lower surface area would also make for lesser environmental sensitivity (such as moisture), and the single massive grain would eliminate the problem of loose powder shifting inside a partially-full shell.
- High-acceleration rockets - A need exists for very high acceleration rockets, such as anti-missile missiles. The only way to achieve such high power outputs within the state of the (solid) propellant art is to use high surface area, as in the gun propellant case above. It would appear that this would require thin, fragile propellant webs with concomitant inefficient volume utilization. A very fast-burning intermediate explosive could permit use of a single, cast, end-burning grain.
- Acceleration of fragile objects - There are classified weapons applications where a need exists to accelerate warheads or warhead components which would be damaged or destroyed by a detonation. There are other classified applications where a detonation will not couple efficiently with the component to be accelerated because of the short duration of the pressure pulse. The gentler, prolonged thrust of an intermediate explosive could solve both problems.
- Bursters for materials which a detonation would destroy - Cannisters containing insecticides, fogs and other (classified) materials often use explosive charges to disperse their contents. One problem is that the hydrodynamic shock of a detonation generates sufficiently high temperatures to decompose some of the materials of interest. The gentler intermediate explosives could serve as non-destructive bursters.
- Demolition - It is well-known that high explosives do not couple well to target structures in near-miss situations, whereas low explosives can do far more damage pound for pound. Intermediate explosives should combine the most effective features of each.

1.3. Outlook for Realization

The prospects for developing stable, reliable, intermediate explosives based upon present explosive art are not extensive; but there are three promising leads.

The McCormick-Selph Company of Hollister, California, claims a family of solid explosives which react at rates of 4.5 to 300 meters per second with pressures below about 2 kilobars. The materials are proprietary, and McCormick-Selph did not wish to reveal their composition at this stage; but they did say that they are based upon a family of pure compounds which are virtually gasless monopropellants and which yield approximately 450 cal/gm. The pure compounds burn at approximately 4.5 m/sec in long, thin columns or in 0.5 inch diameter pressed pellets. The addition of an inorganic oxidizer (up to 85 wt.%) increases the heat yield up to approximately 1500 cal/gm, and the gas yield up to somewhat more than smokeless powder. The propagation rates of oxidized compositions range up to 280 m/sec.

The pressures generated appear to be in the intermediate range. McCormick-Selph reports that fuse cords made from these materials will not initiate PETN, which implies a shock pressure of < 2 kbar. An accidental explosion of 1 pound of oxidized mixture on a 1/4 inch steel plate produced a dent approximately 3 inches deep. Experienced observers felt that black powder (non-detonating) would have produced no dent, and that an ordinary high explosive would have punched a clean hole. Full-scale card-gap tests have not been run; however, initiations of columns and/or pellets by squibs, blasting caps, Primacord, and/or a 1/2 inch thick stack of RDX sheet explosive have always given deflagrations as described above, and never detonations.

The Pyrofuse Corporation of Mount Vernon, New York, reports that bimetallic aluminum-palladium granules will "burn," by alloying, at rates of up to 250 cm/second, when packed into 0.1 inch diameter columns at a density of 3.5 grams/cm³. The reaction is one of alloying, not oxidation; and the reaction product is solid Al-Pd alloy. Conceivably, the addition of an oxidizer, such as ammonium perchlorate, might accelerate the reaction still more, as in the McCormick-Selph case, and produce gaseous working fluid as well.

Dr. M. A. Cook of Intermountain Research and Engineering Company of Salt Lake City, Utah, reports that ammonium nitrate granules, suitably coated with high explosives such as nitroglycerin will detonate at velocities as low as 1100 m/sec - about 1/6 the rate of normal condensed-phase detonations.

Each of these leads is discussed more completely in Sections 2.3.1. and 3.2.1., in the body of this report.

2. STATE OF THE ART OF REACTIVE FLOW PROCESSES

Intermediate explosives can only be understood in the context of the state of the art of explosives in general; consequently, this section presents a review of reaction wave theory brought up to date (1967), followed by a description and interpretation of apparently intermediate phenomena which have been observed.

2.1. Introduction and Summary

The current thermo-hydrodynamic theories of rapid exothermic processes dictate two and only two distinct, stable, combustion regimes, known as "deflagration" and "detonation." Detonation reactions are shock-initiated at the wave front and propagate at supersonic (shock) velocity. Detonation waves produce pressures between one kilobar and one megabar, have short reaction times in the range between a fraction of a microsecond and several microseconds within the wave, and have propagation velocities from one to ten thousand meters per second. Deflagration reactions depend on relatively slow transport processes such as conduction and convection for their propagation and hence are necessarily subsonic. Conventional unconfined deflagration waves generate pressures of approximately one atmosphere, have relatively long reaction times in the millisecond range within the wave, and have propagation velocities of a few centimeters per second.

Deflagration and detonation are similar phenomena in that both processes receive their sustaining energy from exothermic chemical reactions, and many materials are capable of undergoing either reaction regime as well as transition from one regime to the other. The fundamental difference between the processes lies in the initiation and propagation mechanisms of the reaction waves.

Detonation research in condensed-phase systems has been almost entirely based on the von Neumann, Zeldovich, Döring theoretical model. This model contains many simplifications, but correctly describes the essential, observed characteristics of detonations in the great majority of cases. Adequate experimental test of this model for condensed systems has until recently been impossible due to lack of equation of state information; but within the last year, quantitative evidence of its inadequacy to describe all the details of condensed system detonation has been presented (5). Thus, while this theory will always provide an excellent first approximation and useful calculation aid, it can no longer provide a reliable foundation for all aspects of detonation research.

Deflagration of a gas can be accurately described theoretically in terms of thermodynamics, hydrodynamics and kinetics, although sufficient kinetic data are seldom available. Deflagration of solids is more complex and can at present be only qualitatively described via mechanistic models. The essential mechanisms are currently being clarified and will be more accurately determined in the future. Future progress in the field will also be accomplished through more accurate determination of physical and chemical parameters.

The state of understanding of reaction waves which appear to be intermediate between conventional deflagration and conventional detonation can best be described as poor. Current theories do not admit the possibility of stable reaction regimes having propagation rates and pressures intermediate between those of deflagration and detonation. However, for many years, experimenters have observed a variety of combustion phenomena which do not correspond to those predicted by the usual theoretical models, but which propagate at velocities intermediate between conventional deflagration and conventional detonation. They are often, but not always, transient.

2.2. Deflagration and Detonation Processes

Deflagration and detonation are similar processes representing opposite limits of the spectrum of reactive flow phenomena, and the development of understanding of these two phenomena has been complementary. This survey covers an extremely broad area and thus cannot claim to be entirely comprehensive; but an attempt has been made to define those areas of particular significance and cover them to the depth necessary to provide an appreciation of the subject matter for the reader.

A very extensive literature has been drawn upon, but this literature has been specifically referenced only when specific items and ideas from it have been cited. A further bibliography is appended.

Earlier reviews by Gross and Oppenheim (1) concerning gaseous detonation, and by Jacobs (2) concerning condensed phase detonation, cover certain aspects of the subject matter in more detail than does this review. Readers interested in the detailed consideration of theoretical approaches to the phenomena of deflagration and detonation are referred to the excellent reviews of those subjects provided by Evans (3,4).

2.2.1. History of Investigations

The study of chemical reactions of the "combustion" type was stimulated in 1881 by the discovery by Berthelot and Vieille (6) and Mallard and LeChatelier (7) that, under certain conditions, processes having wave characteristics could be observed during experiments related to the combustion of gases in tubes. Berthelot and Vieille measured the propagation velocity of the observed waves and developed a theory to explain the experimental data (8,9). Mallard and LeChatelier noted oscillatory movements of the flame front preceding the transformation from the initial uniform motion to high velocity combustion, and employed photographic techniques, using a rotating drum camera (10).

From 1881 to about 1930, "detonation," the term which came to be applied to the wave process, was studied intensively. During this period, the promoting influence of turbulence was discovered, shock waves preceding the flame front were observed, and various spectroscopic studies were made.

LeChatelier (10) and Dixon (11) measured the detonation induction distance, which they considered to be the prime parameter characterizing detonation. Vieille (12,13) considered gas dynamics and interpreted the observations of others with respect to the possible role played by shock waves on the process. Chapman (14) and Jouguet (15,16,17,18) analyzed the thermodynamic consequences of the "detonation" wave and postulated that a detonation consisted of a shock wave followed by a deflagration. Shock waves had been exhaustively studied by Hugoniot (19) some ten years before.

The work of the next period of years was concentrated on improving the combined thermodynamic-hydrodynamic approach initiated by Chapman and Jouguet. Experimental work involved the measurement of detonation velocities and limits of detonability. Russian investigators during the same time period were interested mainly in the gas dynamic effects and the influence of heat transfer phenomena on chemical reactions (20,21). In this regard, Shchelkin (22) interpreted the role of pre-flame shocks on flame acceleration. He and Zeldovich (23,24,25,26,27,28) attributed most of the observations to the effect of turbulence. Kogarko and Niovkov (29) performed experiments which proved that conditions favorable to the transition to detonation were created when turbulence was formed ahead of the flame front.

Schmidt (30) made an early attempt (1935) to extend the theory of detonation in quantitative terms to condensed explosives. It was not until early in the World War II era, however, that such extension was accomplished. The major efforts were provided by Kistiakowsky (31), Ratner (32), Caldirola (33), Cook (34), and Jones (35). These investigators derived detonation properties utilizing different forms for equations of state for the reaction products, etc. The good agreement between the analytical and experimental results achieved by these experiments left little doubt regarding applicability of the theory.

Basic understanding of the phenomenon of detonation waves was achieved in the World War II era through the work of von Neumann (36), Zeldovich (24), Burkhardt (37), and Döring (37,38). These investigators, utilizing the Chapman-Jouguet idea, described detonation as a reaction zone following a non-reactive shock wave; and it was from this work that modern ideas and experimental techniques developed. Their model contains many simplifications, and neglects dissipative effects entirely, but correctly describes the essential, observed characteristics of detonation in the great majority of cases. Further elaboration and better equation of state data are needed to describe all the details of condensed phase detonation, particularly the temperature and side-loss effects.

2.2.2. Relationship of Deflagration and Detonation

The theoretical descriptions of deflagration and of detonation processes follow from a single line of mathematical reasoning. Courant and Friedrichs (39) have provided the most detailed exposition of the theory underlying the study of reaction waves; and their work, although originally confined to gaseous systems, will be followed here to the extent applicable to condensed phase systems, which are of greater interest in this survey.

In the development of the theory, it is assumed that the chemical reaction which takes place is instantaneous. Thus, a sharp front exists between the unreacted material and the products of reaction. The transition across such a front is analogous to the transition of uncompressed to compressed material across a shock front in a non-reacting material. There does exist a fundamental difference, however, in that the reacted material differs from the unreacted material in specie; and energy has been released in the reaction process, influencing the energy balance.

The mathematical description of the transition from the unreacted to the reacted state is determined through consideration of conservation of mass, momentum, and energy. Observing the one-dimensional process from a coordinate system moving with the front, conservation of mass and momentum yield:

$$\rho_0 v_0 = \rho_1 v_1 = m \quad * \quad (1)$$

$$P_0 + \rho_0 v_0^2 = P_1 + \rho_1 v_1^2 \quad (2)$$

Identical to that obtained for pure shock waves.

Conservation of energy yields

$$E^{(0)}(v_0, P_0) + P_0 v_0^2 + 1/2 v_0^2 = E^{(1)}(v_1, P_1) + P_1 v_1^2 + 1/2 v_1^2 \quad (3)$$

where E represents total energy and is a different function of V and P on the two sides of the reaction front. This differs from pure shock theory in that E represents total energy as opposed to internal energy. The consequences of this difference are significant.

Combining the so-called mechanical conditions, equations (1) and (2), yields

$$(P_1 - P_0)/(v_1 - v_0) = -\rho^2 v^2 = -\rho_0^2 v_0^2 \quad (4)$$

Equation (4) indicates that pressure and specific volume increase or decrease in opposite directions (equivalently, pressure and density increase and decrease in the same direction). It therefore follows that two different types of processes are compatible with the laws of conservation. Processes in which both pressure and density increase are called detonations, and those in which both pressure and density decrease are called deflagrations.

*Symbols used are defined on fold-out page 39.

A characteristic difference between deflagrations and detonations can be seen by rearranging Equation (1) which represents conservation of momentum:

$$(P_1 - P_o)/(U_1 - U_o) = -\rho_o v_o = -\rho_1 v_1 \quad (5)$$

Thus, when pressure increases, in a detonation, the velocity of the products of reaction is less than that of the reactants relative to the reaction front. When the pressure decreases in a deflagration, the products are accelerated away from the reaction front.

Eliminating the velocities from Equations (1), (2), and (3) yields the Hugoniot relation

$$E^{(1)}(v_1, P_1) - E^{(o)}(v_o, P_o) = -1/2 (v_1 - v_o)(P_1 + P_o) \quad (6)$$

Forming the Hugoniot function

$$H^{(1)}(v, P) = E^{(1)}(v, P) - E^{(1)}(v_o, P_o) + \frac{1}{2} (v - v_o)(P + P_o) \quad (7)$$

Thus the Hugoniot relation Equation (6) becomes

$$H^{(1)}(v, P) = E^{(o)}(v_o, P_o) - E^{(1)}(v_o, P_o) \quad (8)$$

For a given v_o and P_o , v_1 and P_1 will satisfy Equation (8) in all reaction processes for which the conservation relations (Equations 1, 2, and 3) are obeyed (The reaction processes must be exothermic). This excludes certain values of v_1 and P_1 in that the condition

$$\frac{(P_1 - P_o)}{(v_1 - v_o)} < 0 \quad (9)$$

from Equation (4) must be satisfied. A plot of the points satisfying Equations (8) and (9) is indicated in Figure 1 (on fold-out page 40). Such a plot is called the Hugoniot curve or Rankine-Hugoniot diagram for the reaction products. The curve is seen to have two branches, exhibiting the compatibility of the conservation laws with two different types of processes. The branches are referred to as detonation and deflagration according to $v \leq v_o$ or $P \leq P_o$. The various possibilities for a reactive flow process beginning at the point (v_o, P_o) can be obtained by considering Figure 1.

Consider a straight line (equivalent to Equation 4) through the initial state point intersecting the deflagration branch of the Hugoniot. If the slope $(P-P_0)/(V-V_0)$ is a small negative number, it intersects the Hugoniot near the point B corresponding to a constant-pressure deflagration. If the slope is decreased, ultimately two intersections with the Hugoniot occur (two is the maximum number which can occur). The first intersection is called a weak deflagration and the second a strong deflagration. These points are separated by C, the Chapman-Jouguet deflagration which is the final point of coalescence of the two intersections as the slope is decreased. This point also corresponds to a maximum in the entropy of the deflagration products as well as in the reaction front velocity.

Similar consideration of a straight line through the initial state point intersecting the detonation branch of the Hugoniot yields, if the slope of the ray is a large negative number, an intersection near point A, corresponding to a constant volume detonation. Decreasing the slope yields two intersections known as weak and strong detonations. The two points of intersection ultimately coalesce at D as the slope is changed. Point D is known as a Chapman-Jouguet detonation. At this point the velocity of the reaction wave and the entropy of the reaction products are at relative minima. The velocity of the reaction products relative to the reaction front is equal to sonic velocity in the reaction products at the C-J point.

Thus, there exist six classes of reaction waves about which general statements can be made. The statements are known collectively as Jouguet's Rule which states that the flow relative to the reaction front is:

- Sonic behind a C-J detonation
- Supersonic ahead of a detonation
- Supersonic behind a weak detonation
- Subsonic behind a strong detonation
- Subsonic ahead of a deflagration
- Subsonic behind a weak deflagration
- Supersonic behind a strong deflagration
- Sonic behind a C-J deflagration

The list enumerates the combinations of processes which are mathematically compatible with the conservation laws, and has no connection with physical reality. Further analysis imposes more stringent limitations with respect to the processes that are physically possible, and the two principal processes are discussed in the following sections.

2.2.3. Detonation Processes

Detonation models beyond the simple theory presented above have had the goal of systematically removing various simplifying assumptions in order to come as close as possible to physical reality. The models have progressed from one-dimensional, steady state flow with instantaneous reaction, to three-dimensional, transient flow with finite reaction rates; but the work beyond that represented by one-dimensional steady state flow has to date been of only limited applicability.

The reactive flow process known as detonation concerns the upper branch of the Hugoniot curve (Figure 1) and involves the postulate that the velocity of the reaction products relative to the wave front is necessarily equal to sonic velocity in the products in order for a stable detonation wave to propagate. The postulate was made by Chapman and, independently, by Jouguet. Brinkley and Kirkwood (40) have advanced a hydrodynamic argument leading to the same conclusion; that is, the Chapman-Jouguet point represents the only possible stable reaction wave in the detonation regime. This is easy to see by making logical application of Jouguet's rule: If a strong detonation (point E, Figure 1, p. 40) were to be initiated, the flow behind the front would be subsonic, and rarefactions would lower the pressure at the front. The velocity of the front would consequently decrease and point E would approach point D. Conversely, if a weak detonation (near point A, Figure 1, p. 40) were to be initiated, the flow behind the front would be supersonic, reinforcing the front and increasing the pressure. Thus, the velocity would increase and point A would move toward point D. It can readily be seen, therefore, that point D represents the point of balance, such that the front is not affected by the flow behind it. The flow is stable under this condition.

A reaction zone of finite width (that is, a finite reaction rate) and the consequent progression of the states through the reaction zone were considered by Zeldovich (23), von Neumann (36), and Döring (30), who independently postulated that a detonation is a combustion process initiated by shock wave. They pictured the shock passage as complete before appreciable reaction took place. The detonation process thus becomes a shock followed by a hydrodynamic deflagration in which the pressure decreases to a value which satisfies steady state requirements (ZND theory). The pressure change is defined by

$$(P - P_0)/(V - V_0) = -\rho v^2 = -\rho_0 v_0^2 \quad (4)$$

the locus of which is known as a "Rayleigh line."

The process can be represented on a Rankine-Hugoniot diagram as indicated in Figure 2 (on fold-out page 41, following). In Figure 2, curve AN represents the Hugoniot function of the reactants, while curve BC represents the Hugoniot function of the products. Line ADN represents a Rayleigh line, point D being the Chapman-Jouguet point. Equation (4) indicates that, if the shock wave and the chemical reactions actually occur separately, the Rayleigh line must in fact be straight. The sequence of events, then, is (1) a shock wave occurs in the reactant material in which the state changes from point A, the initial state, discontinuously to point N, followed by (2) a combustion process in which the conditions move down the Rayleigh line from point N to point D, the Chapman-Jouguet point or point of stability, as the reaction proceeds. Point D represents satisfaction of the conservation relations. The necessary shock discontinuity at the wave front causes a pressure spike in the region between the wave front and the Chapman-Jouguet point, represented by point N. For this reason, the ZND theory has been referred to as the "spike" theory. Point N actually represents the pressure behind a shock wave in a non-reactive material necessary for the shock wave to have a velocity equal to the observed detonation velocity.

The qualitative features of the structure of a detonation wave according to ZND theory are indicated in Figure 3 (on fold-out page 42, following). In Figure 3, pressure, density, and temperature throughout the detonation wave are shown as a function of distance, and the letters on the abscissa are keyed to the points of Figure 2. It can be observed that the pressure at the spike, point N, is approximately twice the pressure at the Chapman-Jouguet point. The temperature is about half the temperature at the Chapman-Jouguet point. Thus, the first phase of the detonation wave consists of an ideal shock in which very little chemical reaction takes place. The second phase is a gradual decrease in pressure and increase in temperature, concurrent with the completion of the chemical reactions. The second phase occurs within a distance of the order of one centimeter for gaseous detonations, significantly less for condensed phase materials. This is the so-called reaction zone length. The ZND theory supplies the logical hydrodynamic link between shocks in non-reactive media and detonations.

Hirschfelder and Curtiss (41) and (42) have employed a more sophisticated approach in order to avoid the shock postulate inherent in the ZND theory. In their work, the equations of conservation of mass, momentum, and energy, including the effects of viscosity, heat conduction and diffusion, along with chemical reaction, were solved numerically for the interior of the detonation wave. Their results indicate that a detonation begins with a shock of finite width and that the Chapman-Jouguet hypothesis is correct for the plane at which the chemical reaction is complete, provided that the viscosity and heat conductivity coefficients are sufficiently small for a given reaction rate. Some chemical reaction occurs within the shock because the shock has finite width. Thus the shock and the reaction zone are coupled and not independent as indicated in the ZND theory. The peak pressure at the wave front represented by point N in Figure 2, can become significantly smaller as a result of this coupling.

A great deal of effort has been put forth over the years to verify the predictions of the Chapman-Jouguet theory experimentally. The theory has been shown to be essentially correct for gaseous detonations although there are notable exceptions, a prime example being spinning detonations (43). The theory is considerably less exact for condensed phase detonations but still furnishes a good first approximation. Lack of adequate information concerning the equation of state of detonation products from condensed phase materials has in general prevented quantitative experimental tests of the Chapman-Jouguet predictions for condensed phase detonations, although Duff and Houston (44) obtained excellent confirmation of the hydrodynamic consequences of ZND Theory for the condensed explosive, Composition B. They determined reaction zone length and C-J pressure, using experimental measurements of the free surface velocity imparted to thin metal plates by the detonation wave.

Wood and Fickett (45) have proposed experiments designed to test the validity of the Chapman-Jouguet theory and which do not require knowledge or assumptions concerning the nature of the equation of state. This technique involves calculation of a Chapman-Jouguet pressure from changes of detonation velocity with respect to initial density and initial energy of the condensed

phase material. Davis, Craig and Ramsey (5) recently reported the results of an experimental investigation utilizing this technique. The initial state of the explosive was varied by using mixtures of nitromethane and another liquid, made of equal molar parts of nitric acid, acetonitrile and water, which had the same atomic composition as nitromethane. In another example, they varied the initial state by using TNT as a liquid and as a solid. Calculated Chapman-Jouguet pressures were compared with directly measured pressures; and the two differed by 15-20%, a disagreement too large to be the result of systematic error in the measurements. This is felt to be conclusive proof that Chapman-Jouguet theory does not agree with experiment for nitromethane, solid TNT and liquid TNT. The authors offer no replacement for the theory and only speculate about the failure. They suggest that the failure may result from very rapid oscillations in the flow.

Erpenbeck (46,47) has mathematically investigated the stability of the steady-state detonation equations and has found numerous cases of instability. Because of ignorance of the details of reaction processes in liquid and solid materials and of the equations of state, it is not possible to determine whether this instability can be expected in real explosives. Wood and Fickett (48) and Mader (49) have made numerical calculations of the mathematical behavior of various detonation models and have found instability which manifests itself as a periodic variation of the flow. Mader found very large oscillations in studying the reaction zones of nitromethane and TNT. His work employed very accurately determined physical/chemical parameters. This suggests that the familiar assumptions of stable, one-dimensional detonation are not valid in any system when looking at small-scale details.

Thus, the basic assumptions of the von Neumann, Zeldovich, Döring elaboration of the Chapman-Jouguet theory appear to be invalid on a micro-scale; and the theory as a whole can no longer be accepted as a sufficient description of the detonation process. It, however, provides an excellent first approximation and a very useful aid in detonation calculations. White (50) has initiated the development of a turbulent detonation theory for gaseous systems. It is possible that this theory can serve as the basis for the development of a more satisfactory detonation theory applicable to condensed phase materials. When this occurs, we will be in a better position to appreciate the limitations of the present theory.

One of the major limitations of the present detonation theory is that it describes systems that are essentially infinite in size, i.e., an increase in size will not change the characteristic velocity and pressure of the detonation. Real problems, however, often involve systems small enough so that the velocity and pressure are affected by changes in diameter. Detonations occurring in such systems are generally known as "nonideal detonations."

The movement of mass and energy along coordinates other than those normal to the detonation front is not considered in one-dimensional steady state detonation theory, and modified theoretical models have been developed to describe such non-ideal detonations. Several models have been proposed. These include the "nozzle" theory of Jones (51), the "curved front" theory of Eyring, et al. (52), and the "geometrical model" of Cook, et al. (53). The first two of these theories have in common the assumption that only the equation of continuity is perturbed by the radial expansion of the reaction zone. The latter theory treats "effective" reaction zone length as a function of lateral losses. Of these theories, the nozzle theory and geometrical model appear to give the best agreement with experimental data. Experimental data (54) confirm that detonation fronts are indeed curved under conditions of non-ideal detonation. The relationship between detonation velocity and curvature of the detonation front has been explored theoretically by Wood and Kirkwood (55). These theories are discussed more fully in Section 2.3.2. of this study.

2.2.4. Deflagration Processes

The reaction flow process known as deflagration concerns the lower branch of the Hugoniot Curve (Figure 1), but not any single point on it. A set of deflagration expressions exists for any given initial state, and all members of the set satisfy the conservation conditions. Thus, deflagration processes possess a higher degree of indeterminacy than detonation processes, but consideration of the internal reaction mechanisms makes it possible to exclude certain classes of deflagration waves. Courant and Friedrichs (39) have rigorously shown that strong deflagrations are never possible and that weak deflagrations are possible only with a well-determined velocity, depending on the state of the unburnt fuel and on the thermal conductivity. Thus, deflagration differs significantly from detonation in that the internal reaction mechanisms of chemical kinetics and the transport processes of diffusion and thermal conduction control the deflagration velocity. Another primary difference lies in the fact that deflagration takes place with a drop in pressure in the reaction products as they expand, a property sometimes considered to be the chief characteristic of deflagration.

Theoretical consideration of deflagration is plagued by two types of difficulties. The first concerns the lack of detailed knowledge concerning chemical kinetic processes, and the second concerns the formidable mathematical equations which result from using the inadequate kinetic theories which are available.

Reaction kinetics constitute a significant problem, because the rate of reaction is a function not only of the temperature, but also of the concentrations of the individual components or species taking part in the reaction. Since many reactions take place simultaneously, intermediates as well as original reactants must become a part of the formulation. Complete experimental data of this type are almost never available. A further complication arises from the fact that the rate dependence on temperature and species concentration can vary significantly from one reaction to another. This fact essentially eliminates the possibility of a general formulation of the problem.

A great many attacks on these problems have been proposed, using various simplifications, and some of these have been rather successful in gaseous systems. Theories of condensed phase deflagrations are in a considerably less satisfactory state.

Hirschfelder and Curtiss, in a series of papers (56,57,58), set up the general, one-dimensional, steady state, constant pressure, flame equations to be solved by computer. The equations formulated were completely general with respect to the number of reaction steps, the number of reactants, and the order of the reactions. Also employed were diffusion equations formulated in terms of the composition gradients. While this technique offers excellent results, the difficulties in employing it and the amount of computation time required make general application prohibitive.

Linnitt (59) has pointed out that there exist two primary mechanisms which limit deflagration velocities. The first of these is thermal conduction, and the second is diffusion. The so-called thermal theories consider heat conduction to be the physical process which limits the rate at which energy can be transferred from the reaction zone forward to fresh layers of fuel. In this manner, the temperature of the unburnt fuel is raised to a point sufficient for the reaction rate to become significant. The diffusional theories postulate that active radicals produced by the chemical reaction diffuse forward into the fresh fuel and initiate reaction. There are, of course, a great many cases to be considered among the diffusional theories as a result of the various types of reaction processes which can occur, each producing radicals with different characteristics.

Among the thermal theories, those of Crussard (60), Damk8hler (61), Daniell (62), Jouguet and Crussard (63), and Nusselt (64) while having somewhat different assumptions inherent in their development, have as a unifying characteristic the fact that the flame velocity varies as a function of the same grouping of parameters including thermal diffusivity and temperature differences. Experimental attempts to verify theories such as these have led to various conclusions, depending upon assumptions regarding such unknown factors as thermal conductivities, etc.

Emmons, Harr, and Strong (65) assumed a first order reaction of the Arrhenius type; and using constant physical/chemical parameters, including energy release, solved the resulting energy equation numerically. They determined deflagration velocity as a function of temperature gradient, and found that a most probable deflagration velocity exists for mixtures having high heat release rates. For mixtures with low heat release rates, they showed that any deflagration velocity could exist.

Bechert (66) formulated a thermal type theory wherein diffusion processes were taken into account in that the activation energy used in the rate expression was determined empirically from experimental data taken under conditions in which diffusion was present. Comparison of the Bechert theory with experimental data indicates that the flame velocity was predicted to the proper order of magnitude and that its dependence upon compositional parameters was qualitatively correct.

Among the diffusional theories, the theory of Tanford and Pease (67, 68) considers that the deflagration velocity is determined by the rate of diffusion of active reaction centers forward into the unburnt fuel. Tanford and Pease developed an equation expressing deflagration velocity in carbon monoxide as a function of diffusion-controlled production of active atoms. A similar equation was developed for expressing the deflagration of hydrogen. Their equations enable calculation of deflagration velocities with errors of not more than 25% in such systems.

Van Tiggelen (69) proposed a theory emphasizing chain branching as the reaction mechanism and developed an expression for the deflagration velocity, assuming the velocity to be limited by the rate of chemical reaction. Calculated deflagration velocities in methane/air mixtures agree well with experimental values obtained by Coward and Hartwell (70).

Other diffusional theories, employing other mechanistic approaches, have been developed by Gaydon and Wolfhard (71,72), Bartholome (73), and Manson (74). All of these theories have the calculation of deflagration velocity as their objective, and none are particularly successful in achieving this goal.

A third group of theories has been developed which are more comprehensive than the formerly discussed groups in that they consider both the conduction of energy and diffusional processes in the calculation of deflagration rates.

Lewis and von Elbe (75) considered a simple, exothermic ozone-to-oxygen reaction and measured flame velocities at various pressures and initial temperatures. They considered the effect of heat conduction as well as diffusional processes, and included the equations of continuity in their development. In addition, they explicitly introduced reaction rate as a function of concentration and temperature. Their calculated flame velocities were of the same order of magnitude as those observed.

Corner and Boys (76,77) utilized simplified reaction scheme models (a single exothermic reaction) and a method of successive approximations which allowed analytical solutions to be obtained. Numerical solutions of the equations were compared with the approximate analytical methods, and it was found that the analytical approximations worked quite well.

Zeldovich and Frank-Kamenetskii (78,79) employed a more complicated reaction model in the development of a theory for which the order of reaction may be zero, first, or second with respect to the reactant. The result of this theory is a series of formulas describing the cases of unimolecular, bimolecular, etc., reactions, of each order; and Seminov (80) has provided estimates of the range of applicability to which the various formulas apply. The Frank-Kamenetski, Zeldovich theory has been employed by a number of other investigators (81,82,83,84) with generally quite good results.

von Karman and Penner (85) formulated the complete system of equations expressing laminar flame theory, including diffusion and heat conduction. They considered an arbitrary number of simultaneous reactions. Explicit, approximate formulations were developed for the flame velocity without the employment of physically unrealistic assumptions. Examples of hydrazine decomposition and ozone decomposition show good agreement between the approximate theory and detailed numerical integration of the complete equation system. The theory develops a more complete description of the general area of chain reactions, offering indications that more complicated chain reaction problems can be solved. However, the work of Herschfelder and Curtiss and that of von Karman has brought the theory of laminar deflagration to full maturity. It is now possible to obtain a complete chemical and physical picture of the composition of the reaction zone for any given set of physical and chemical parameters. This becomes a useful tool in the study of the kinetics of combustion processes.

All of the above discussion pertains to laminar deflagration in gaseous systems; however, combustion reactions are generally accompanied by the evolution of thermal energy which causes motion in the unburned fuel ahead of the flame front, so that the combustion becomes unsteady, rapidly developing into an accelerating or vibrating flame. Since "deflagration" strictly includes only regimes of constant velocity flame propagation, described by the lower branch of the Hugoniot curve; accelerating or vibrating flames, which do not meet the steady flow conditions, cannot in general be treated using deflagration theory. Many investigators have attempted to relate accelerating flames to the Hugoniot curve by assuming that the state of the combustion products is represented by a point which shifts along the Hugoniot curve when the flame velocity varies. In reality this assumption is incorrect. Deflagration theory, as such, is not applicable to turbulent combustion or to the combustion of condensed phase materials.

Most theories of solid combustion consider that two consecutive reaction steps take place; the first producing a phase change, and the second producing exothermic combustion. It is necessary for the solid material to become vaporized before the combustion reaction can begin. Thus, the burning rate of such materials is in general limited by the rate of surface vaporization which in turn is determined by the rate at which heat is transferred from the hot, gaseous reaction products to the unreacted solid material. The reaction zone is almost vanishingly thin because the very steep temperature gradient is reflected in an ever steeper reaction rate gradient.

The rate at which heat is transferred from the gaseous reaction products to the solid reactant is the product of the difference between their temperatures and the coefficient of heat transfer. The heat transfer coefficient is a function of the flow conditions of the gas and its thermodynamic properties, and is directly proportional to pressure. Thus, the rate at which heat is transferred from the products to the reactant, and consequently the surface burning rate, should be directly proportional to pressure. In actuality, it is found that the increase in surface burning rate with increasing pressure is somewhat less than linear. This is the

result of the fact that the heat of vaporization of solid materials usually increases with increasing ambient pressure. In addition to the effect of pressure, it has been found that linear burning rates of condensed phase materials increase with the temperature of the solid material and increase with the velocity of the combustion gases parallel to the burning surface.

The effects of pressure and gas velocity were first observed and discussed by Muraour (86,87) in his extensive studies of explosive powders and gun propellants. The effect of temperature on burning rate has been described by Reid (88) and Geckler and Springer (89) who have proposed empirical equations useful for estimating the magnitude and direction of the effect.

Recent work has been stimulated by the interest in solid rocket propellants, and a relatively large effort has been put forth in the development of theories of combustion for condensed phase materials over the last 15 years. Important contributions have been made by Wilfong, Penner, and Daniels (90), Parr and Crawford (91), Rice and Ginnell (92), Summerfield (93), Friedman (94), and Corner (95).

In the Wilfong, Penner and Daniels work (90), it was proposed that the controlling step in the process of condensed phase combustion is a solid-phase chemical reaction at the propellant surface and that ambient pressure does not affect the rate of the solid phase reaction. However, experimental burning rate data indicate that pressure has a strong effect.

The procedure used by Rice and Ginnell (92) is based upon observations of the flame zone in composite propellants by Crawford (96), which divides the gaseous region into several arbitrary zones. It is proposed that the burning rate is controlled by a combination of the kinetics of an initial solid phase reaction, the heat transfer rate to the solid by conduction, the combustion kinetics in the gas phase, and diffusion from the surface to the bulk gas. Application of the theoretical equations indicated the proper increase of burning rate with pressure, and the magnitude of this effect was found to be dependent upon the order of the gas phase reaction. The final expressions developed in this theory are difficult and cumbersome to use.

The concept of dividing the combustion region into several arbitrary zones was also employed by Parr and Crawford (91). They, however, neglected diffusion in their formulation; and the results of their work do not lead to particularly useful expressions. Summerfield and co-workers experimentally observed the burning region in ammonium perchlorate. They observed a thin gaseous reaction zone, from which it was concluded that no chemical reactions occurred at the solid/gas interface. Thus, it was postulated that the burning surface vaporizes and diffuses into the bulk gas unmixed and unreacted. The theory developed in this work presumed the rate to be controlled by gaseous combustion kinetics and by diffusion and heat transfer near the propellant surface. Empirical expressions for the burning rate were obtained for two cases: the first considered that a second order combustion reaction controlled the burning rate, and the second assumed that the combustion reactions occurred at near-equilibrium conditions (93).

Further development of the theory of condensed phase combustion has been analogous to that in the past in that the two-phase scheme has been employed; but different reaction models and different controlling mechanisms have been proposed. In general, the experimentally observed burning rates can be accurately reproduced by the models which have been developed; but most models can serve only as correlating equations, since the detailed and exacting experimental studies of the intermediate stages in the overall process of combustion have not yet been accomplished, and the actual mechanisms have not been accurately identified.

2.2.5. Equations of State

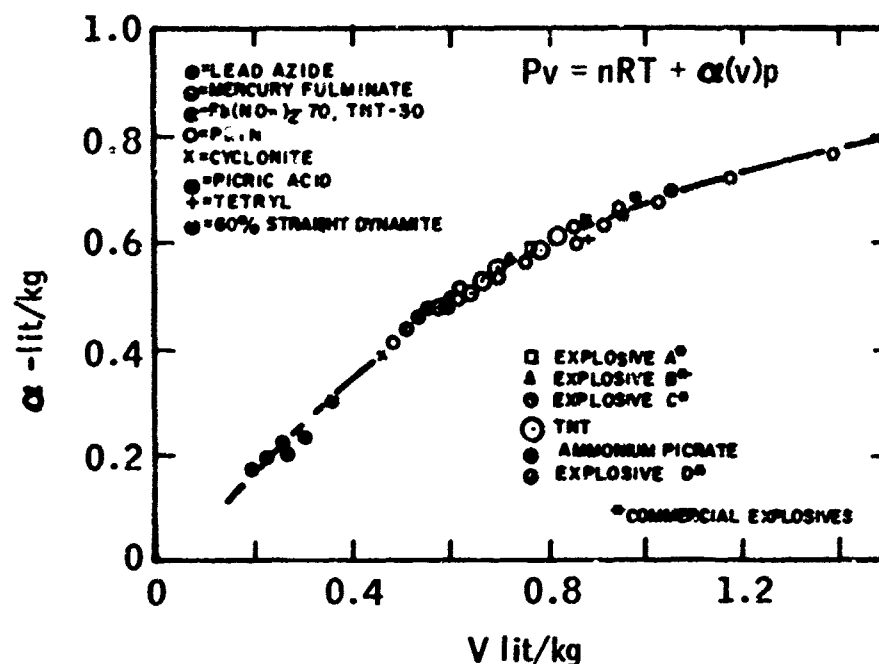
The detonation literature contains a great variety of computations pertaining to the properties of detonation products, and the results and the differences among them are highly dependent upon the form of the equation of state (the relationship among pressure, volume, and energy) selected for the computation. This is especially true of thermal computations; the form of the equation of state makes much less difference in calculating detonation velocity.

Early work with gases stressed the use of the perfect gas law, but this procedure was inadequate for the dense states created by detonations in liquid or solid phase materials; therefore, variations were introduced.

The first studies employed the Abel equation of state (97) which corrects for deviations from the ideal gas law which are the result of finite molecular volume. The Abel equation assumes that the molecular covolume is constant; but work by Langweiler (98) and others, using detonation data to calculate the covolume, showed it to be a function of both temperature and initial density. Thus, since the covolume term assumes overriding importance in the states of interest, the early studies were of little practical value.

Cook (53,99) used density/detonation velocity data to determine covolumes as a function only of volume, assuming that temperature would have only a small effect. This result was relatively successful, and the covolumes for many substances are presented as a function of volume in Figure 4, on the following page. Cowan and Fickett (100) have attacked this procedure on the basis that the equation does not provide for any potential energy of molecular interaction, and thus cannot be accurate at high density. The error should result in the calculation of detonation temperatures which are too high, even though calculated velocities are in agreement with experiment.

Figure 4
COVOLUME AS A FUNCTION OF VOLUME



After Cook (53)

The covolume-as-a-function-of-volume approximation has been investigated by many others including Caldirola, Murgai, Patterson, and Morris and Thomas (101,102,103,104). The latter investigators used a virial form presented by Hirschfelder and Roseveare (105) which yielded the variable covolume as a function of one semi-empirical constant, the van der Waals covolume. This work provided excellent agreement with experiment for explosives at moderate densities.

Jones assumed the covolume to be a function of pressure, and using a quadratic expression for the functionality, computed an isentrope from the Chapman-Jouguet point for TNT (106); and Kistiakowsky, Brinkley, and Wilson (107,108) provided many calculations which expressed the covolume as a function of both volume and temperature, as it should be theoretically. Semi-empirical constants used in calculating covolumes were determined by using those values which provided the best agreement between measured and calculated detonation velocities. This work was extended by Cowan and Fickett (100) who determined new parameters in agreement with the new extensive experimental data which had become available. Their results were tested extensively against new experimental data by Mader, who found that they could not be significantly improved. Mader computed the detonation properties of most condensed phase explosives (109). His calculations include the Chapman-Jouguet state, the Hugoniot for the detonation products, and the isentrope through the C-J point. Mader's results have been employed in many hydrodynamic calculations with excellent results.

Jones (35) has derived an equation combining the Hugoniot equation, Rayleigh line, and the fact that detonation velocity has been found to be a function of loading density, which accurately estimates unknown detonation velocities and particle velocity. The equation contains a constant which includes all of the thermodynamic variables; Jones estimates the value of this constant at 0.25. The use of such a simplification essentially avoids the necessity for an equation of state, but of course can only be employed in highly approximate work.

A number of attempts have been made to employ equations of state which are more satisfying to solid-state physicists, i.e., equations such as the Lennard-Jones-Devonshire equation. Studies by Kihara and Hikita (110), Paterson (111), and Murgai (112) provided rather disappointing results, in that agreement of calculations with experiment was far inferior to that achieved with other less sophisticated equations of state. Somewhat better results were obtained by Fickett, Wood, and Salsburg (113), who used detonation velocity data to determine average pair potentials for the products resulting from the detonation of RDX.

Deal (114) determined the Hugoniot of the reaction products near the Chapman-Jouguet state and obtained the isentrope below the C-J state by measuring the hydrodynamic state of inert materials adjacent to an interface with detonating Composition B. The data were found to fit a polytropic equation of state ($P/\rho^\gamma = K$ for the isentrope) quite well. Fickett and Wood (115) determined an equation of state, using Deals' data, by assuming that the internal energy is a linear function of pressure in the region of interest, and using the simple gamma law representation of the isentrope. They point out that such an equation of state is unsuitable for calculation of thermal behavior, but reproduces experimental detonation velocities quite well.

Other investigators (116) have also shown the applicability of a polytropic gas equation of state for representing the adiabat below the C-J state. These experiments have all been performed at pressures above about 100 kilobars.

Skidmore and Hart (117) have tested the validity of the "gamma law" equation of state at states above the C-J point in Composition B. This was accomplished by determining the hydrodynamic properties of overdriven detonation waves using impedance matching experiments. Pressures up to twice the C-J pressure were generated by flying plate impact. The results of this work confirm the applicability of the constant gamma polytropic equation above the C-J state using the exponent (gamma) appropriate to the C-J state.

Wilkins (118), in attempting to use the constant gamma polytropic equation in hydrodynamic calculations, found that predicted energy transfers to spherical shells were higher than experimental values. He introduced a different form for the adiabat which dipped below the constant gamma form (straight line on log-log plot in P,V space) at 150 kilobars and recrossed the line at 3 kilobars. Thus, less energy is available within that pressure range, implying that the detonation products act more like normal gases at low pressures. On theoretical grounds, Jones and Miller (106), had predicted this form for the adiabat of TNT some fifteen years earlier.

Allan and Lambourn (119) extended impedance matching techniques to the lower pressure regime by employing matching with compressed gases. In this way, they covered the range from 0.2 to 30 kilobars for two different HMX compositions. A modified form of the Wilkins equation was employed to fit the data. The results correctly predicted energy transfer in the Wilkins sphere expansion test.

An additional modification to the Wilkins equation of state has been made by Kury (120), et al., at Lawrence Radiation Laboratory. This work used hydrodynamic calculations pertaining to an expanding cylinder as a test of the equation of state. The behavior deduced from this test could not be correctly described using the gamma law, Becker-Kistiakowsky-Wilson equation, or Fickett's (121) form of the Lennard-Jones-Devonshire equation. The new modification to the Wilkins equation covering the range from 500 kilobars down to 1 kilobar permits extremely accurate calculation of the energy delivered to metals by explosives.

Thus, it is apparent that isentropes from about twice the C-J pressure down to approximately 150 kilobars can be accurately represented by a simple, constant gamma, polytropic equation of state. Consideration of lower pressures requires the use of more complex equations. This is particularly true in those cases in which equations of state are being used in hydrodynamic calculations for predicting the acceleration of metals. The more satisfying (from a theoretical standpoint) LJD type equations do not seem to do as good a predictive job as the empirical representations. The form of the equation of state makes little difference in calculating detonation velocity, but becomes a significant factor when thermal computations are made.

2.2.6. Initiation and Growth to Detonation

The formation of a detonation wave in a material capable of reacting exothermically can be divided into four stages of development:

- Initiation
- Deflagration
- Transition (DDT)
- Detonation

The first stage, initiation, depends upon an external energy source in that an insufficient quantity of energy is being released to sustain the reaction; although, with high-energy explosives, the external energy required can be extremely small. The second stage, deflagration, is a steady state, self-sustaining process following initiation. Energy transmission to unreacted fuel elements takes place via thermal diffusion or other transport mechanisms, and reaction kinetics is not the only rate-controlling factor. Experimentally, it has been found that linear deflagration rates can be described as a function of ambient pressure; thus, as pressure rises, so does deflagration rate -

which leads to the third stage, transition to detonation. In the deflagration to detonation transition (DDT), the mechanisms of energy transmission to unreacted layers of fuel changes from slow transport processes to supersonic shock waves. Detonation, the fourth stage, is one in which the chemical energy is transmitted by shock waves. Detonation, like deflagration, is a steady state, self-sustaining phenomenon. With strong initiations, the deflagration and transition steps may be vanishingly brief or effectively absent; with weak initiations, they can be quite prolonged, even to the point where detonation does not occur at all.

Initiation of detonation, then, can be treated as a shock phenomenon, since detonation is shock-initiated; and one can consider the problem either from the standpoint of what happens when a shock is put into a material, or from the standpoint of the generation of a shock wave as outlined above. This enables separation of problems with respect to the shock source (internal or external); and as far as detonation is concerned, it becomes necessary only to describe the shock duration and intensity. Most theoretical and experimental investigations have started with a donated shock, and studies of shock growth from a small stimulus have received much attention.

Shock-Initiation of Detonation

Most of the work studying detonation initiation from a donated shock has employed some sort of "gap" test. Typically, such tests employ an explosive donor, which sends a shock through a variable-thickness inert barrier into an "acceptor," the explosive material being tested. A series of tests determines a critical thickness of the inert barrier above which detonation is not initiated in the acceptor. Such a test was first suggested by Murawski (122) who had the idea of relating initiation to shock intensity. Later work was accomplished by Cachia and Whitbread (123) and Hertzberg and Walker (124). The gap test provides a rather sharp cutoff between propagation and failure as the result of interactions between reaction rates and hydrodynamics. The shock intensity at the interface between the acceptor and the inert barrier determines the rate of the chemical reaction initiated; and the chemical reaction sends compression waves ahead into the acceptor, strengthening the shock. If the initial shock is strong enough, and if it is sufficiently strengthened within the confines of the experiment, detonation occurs. In general, such testing assumes that initiation is a function only of the peak pressure of the primary shock wave. The duration and amplitude variations of the wave are ignored. Nonplanarity and edge effects are also neglected, as are interactions between the primary shock and the confinement of the acceptor. Such simplifying postulates are not strictly valid and can lead to misinterpretation of results; however, in practice they work fairly well because the neglected parameters are reasonably constant within any given series of experiments.

Another approach to the problem of studying detonation initiation consists of techniques known as "shooting" tests, in which an inert projectile is fired into the material under study. This type of test enables more accurate determination of the donated shock pulse parameters than does a "gap" or

"barrier" type of test, but the advantage is largely offset by experimental difficulties. Griffiths, Laidler, and Spooner (125) used the projectile impact technique and determined the critical projectile velocity for initiation of a series of pressed and cast explosives and determined the effects of various experimental parameters. The explosives were then ranked in descending order of sensitivity.

Brown and Whitbread (126) studied the relationship between "gap" and "shooting" tests, and found a linear relationship between the critical length of brass gap attenuators and the critical velocity of steel projectiles. This relationship was tested and confirmed by Lundborg (127,128,129) for cast TNT, pressed TNT, and pressed PETN.

Gittings (130) used short duration pulses produced by impacting thin films of aluminum on PBX 9404 to study initiation. This method is useful but leads to significant data scatter.

Initiation by impact at velocities possible in accident situations (below 300 m/sec) have been studied by Napader-sky (131) and by Green and Dorrough (132). The phenomena observed varied in severity from high order detonation to mild burning. This work indicates that friction at interfaces and growth of burning to detonation are the factors of greatest importance. The fracture properties of the explosive material strongly affect interface friction. The subject of initiation by mild stimuli has been treated extensively in England under the leadership of Bowden and others at Cambridge (133,134) and is discussed more fully in the following section on "Deflagration-Detonation Transition."

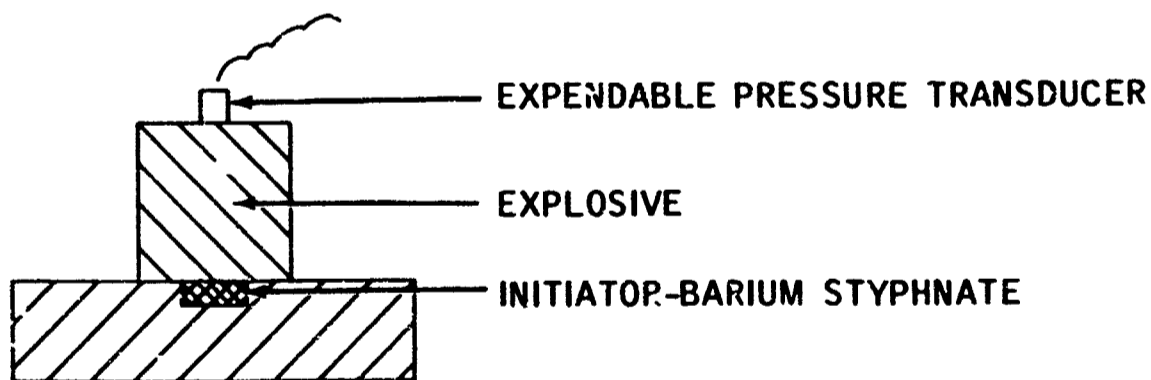
Campbell, Davis, and Travis (135,136) have studied shock initiation in both solid and liquid explosives, using nearly planar incident shock waves, having pressures of 50 to 100 kilobars, from composition B plane wave generators. Experiments with nitromethane, molten TNT, molten DINA, ditherite 13, and PETN have clearly demonstrated the course of events in homogeneous liquid explosives: the explosive is heated by the entering shock wave; and, after an induction period, detonation begins at the donor-acceptor interface, where the material has been at an elevated temperature for the longest time. The resulting detonation wave proceeds through the compressed explosive at a high velocity, overtaking the initial shock and overdriving the unshocked explosive. Experiments with solid explosives, including cyclotol B, TNT, HMX and mixtures of nitromethane-carborundum showed somewhat different results. With such materials the initial wave was found to travel as a nondetonative shock to which the explosive nevertheless contributed energy. The shock velocity therefore increased with time for distances of the order of one cm., after which an abrupt transition to high velocity detonation occurred in approximately 0.01 cm. The transition occurred with no overshoot in velocity as observed with the liquid explosives.

This work demonstrated that initiation is much more dependent upon the detailed structure of the shock properties of the material than upon thermochemical constants, and that homogeneous materials are much more

sensitive in their initiation characteristics to initial temperature and variations of shock pressure than are inhomogeneous materials. However, the degree of homogeneity has a strong effect on the mechanism of initiation, and a shock too weak to initiate a homogeneous explosive can activate hot spots and cause detonation when an explosive is sufficiently inhomogeneous.

Liddiard and Jacobs (207) have completed an experimental study of initiation of reaction in cast explosives. This study covers the borderline region of shock pressures between the point where transition to detonation occurs and the point where no reaction can be detected. The method employed high speed photography and an extension of gap type tests. Results include the pressure at which detonation is just produced. The threshold pressures so determined were found to be lower than those which result from standard gap testing. Liddiard (208) in a more recent study, has subjected explosives to spherical shocks while submerged in water. The threshold for burning was determined to be about one-third of that obtained in the modified gap test. The lower threshold was attributed to increased pulse duration, increased degree of wave planarity and the additional confinement offered by the submergence.

Cachia, working at the Atomic Weapons Research Establishment in England, has devised a similar experiment with which he plans to study low level initiation (209). A diagram of the proposed experimental apparatus is shown below. The initiator provides a pressure pulse which is detected by the transducer. The pressure pulse can be varied over a wide range using this technique.



Muller, Moore, and Bernstein (137) studied the growth of explosion in electrically initiated RDX, using both powdered and pressed RDX. Powdered RDX initiated with an exploding bridgewire was found to support a low velocity detonation wave with abrupt transition to high order detonation. The transition was found to occur at a reproducible induction distance, dependent upon the geometry and loading density. The pressed RDX results indicate that the growth of explosion is governed initially by thermal mechanisms; long induction times of more than a millisecond were noted. Conclusions of these studies can be summarized as follows:

- The initial shock wave in inhomogeneous materials accelerates throughout its travel; the corresponding wave in homogeneous materials attenuates slightly with time.
- The transition to detonation is much more abrupt in homogeneous than in inhomogeneous explosives.
- An overshoot in the velocity of detonation occurs with transition in homogeneous materials; no such overshoot occurs with inhomogeneous materials.
- Detonation originates near the shock front in inhomogeneous explosives and at the donor-acceptor interface in homogeneous explosives.
- Mixtures (e.g., nitromethane-carborundum) are much more sensitive than homogeneous materials. This is probably due to shock interactions which result in local heating.

This work once again demonstrates the dependence of initiation mechanism upon degree of homogeneity.

The advent of high speed digital computers has enabled numerical solution of the equations of hydrodynamics coupled with those of reaction kinetics. Hubbard and Johnson (138) used several reaction rate expressions coupled with one-dimensional hydrodynamic equations to describe the effect of initiating shocks. This work mathematically reproduced the observed phenomena of overshoot in velocity, of buildup, and of failure; and a more complete study undertaken by Boyer, Schermerhorn, and Uyehara (139) showed similar results for the more realistic axisymmetric systems; however, neither of these studies gave results which agreed quantitatively with experimental data. This may have been the result of lack of adequate kinetic and equation of state data.

Mader, in a series of papers (140,141,142,143,144,145,146,147,148) has elegantly addressed the general problem of mathematical simulation of reactive flow. This work has progressed from shock initiation of homogeneous material to hot-spot initiation of materials with varying degrees of inhomogeneity. Agreement with experimental results for nitromethane, liquid TNT, and single-crystal PETN was obtained for homogeneous shock initiation; and it was quantitatively shown in a one-dimensional study that explosion occurs at the donor-acceptor interface as a result of shock heating, i.e., the position of the explosion is that point where chemical decomposition has been occurring the longest. A detonation wave overtakes the initiating shock and initially overdrives the detonation. The introduction of inhomogeneities (hypothetical) enabled detailed explanation of the mechanism of initiation of detonation at hot spots created by shock interaction. The energy transfer mechanism was shown to be shock waves and rarefactions.

In later work, the basic processes of shock initiation of inhomogeneous explosives have been investigated, using a numerical model of a cylinder of nitromethane containing a void or an aluminum pellet. The interaction of shocks with the density discontinuities, the formation of hot spots, and the buildup to propagating detonation were computed using a two-dimensional reactive hydrodynamic model. This work only applies to cases in which the mechanism of initiation failure does not depend on the details of the structure of the reaction zone, as such details were not numerically reproduced.

Details of reaction zone structure in nitromethane and liquid TNT were investigated with a one-dimensional model using Arrhenius kinetics. This study indicated the occurrence of unstable pulsating detonations until the explosives were sufficiently overdriven after which stabilization occurred. The amount of overdrive necessary was found to be primarily dependent upon activation energy.

The latest work in this series treats the shock interactions formed in nitromethane by corners (as in a container) of Plexiglas, aluminum, and gold. The formation of hot spots, with isotherms and the buildup of propagating detonation, has been computed. Excellent agreement has been obtained with experimental results obtained using radiographic techniques. Further studies will include systems with more than one discontinuity and the resulting shock interactions (148). Travis (149) has introduced density discontinuities into nitromethane using various materials, and measured induction times for the initiation of detonation from a plane shock wave. He found the initiating "efficiency" to be strongly dependent upon the material chosen. Geometric factors were also important. These experiments were designed to test the computations of Mader, and experimental results are in excellent agreement with Mader's theoretical results.

The role of cavities in the explosion of liquid materials has been investigated by Watson and Gibson (150) at the U.S. Bureau of Mines, and by Bowden (151) at Cambridge. The work of Watson and Gibson is associated with their study of low velocity detonation, which will be discussed in detail in Section 2.3 of this report. Bowden and co-workers spread nitroglycerine on a flat glass anvil and struck it with a glass hammer after introducing an air bubble. High speed photography was employed to observe the cavity compression and reaction initiation. It was concluded that the formation of micro-Munro jets was an important contributor to the initiation mechanism. This had previously been suggested by Bowden (152) and others.

Price and Petrone (153) have employed the one-dimensional numerical treatment of reactive hydrodynamics constructed by Enig and Metcalf (154) to study the gas loading of explosives to initiating pressure. Deflagration-detonation transition experiments are examples of initiation caused by rapid gas loading. The numerical treatment reproduced the trends found experimentally.

Ablow, Rajapakse, and Evans (155) have analyzed the role of gas compression and conduction in the shock initiation of granular explosives. A theoretical model of an explosive sphere contained within a spherical shell of hot gas was developed. Temperature was computed as a function of time for the surface of the explosive. The results were correlated in terms of dimensionless parameters. The results indicate that the compression-conduction mechanism is unlikely at low pressures, but that it can contribute at pressures of the order of one atmosphere. In practice, granular explosives can be shock initiated independent of ambient pressure; therefore other mechanisms must be operative.

The theoretical ideas concerning the role of interstitial gas in the initiation of low density, granulated, explosives have been examined by Chick (156). This work indicates that interstitial gas lessens the chance for initiation by decreasing convection or by cooling the grains. In addition, this study indicates that initiation can occur as a result of continuous intensification of the entering shock or by an intermediate burning phase which can last for several microseconds. This, of course, has been postulated for some time.

Evans, Harlow, and Meixner (157) have studied the details of shock and rarefaction interactions with a bubble. Numerical techniques for solving the equations of compressible flow were employed, yielding detailed configurations and isotherms for times late in the interaction period. The results for helium bubbles in neon agree qualitatively with experimental data for butane-air.

Seely and Seay investigated the initiation characteristics of low-density PETN (158). This work used nearly-planar shock waves donated through brass and lucite plates to wedge-shaped pressings of PETN. Interstitial voids were filled with gases of widely varying thermodynamic characteristics to evaluate the mechanism of grain burning caused by compression heating of the gas. The point (depth) of detonation initiation was found to be a constant, not a function of the interstitial gas temperature; and initiation was obtained at pressures as low as 2-1/2 kilobars, much lower than the 50 kilobars required for initiation of homogeneous PETN. It is possible that the different temperatures produced by the use of different interstitial gases under compression are compensated by changes in heat transfer efficiency, and this point must be investigated before definitive conclusions can be drawn.

Evans, Reese, Seely, and Lee (159) have studied the shock initiation properties of low density granular ammonium perchlorate by means of extensive experimental work. Initiation of the detonation reaction was found to occur at very low pressures as compared to single ammonium perchlorate crystals. Removal of interstitial gas in a vacuum system had no effect on initiation properties.

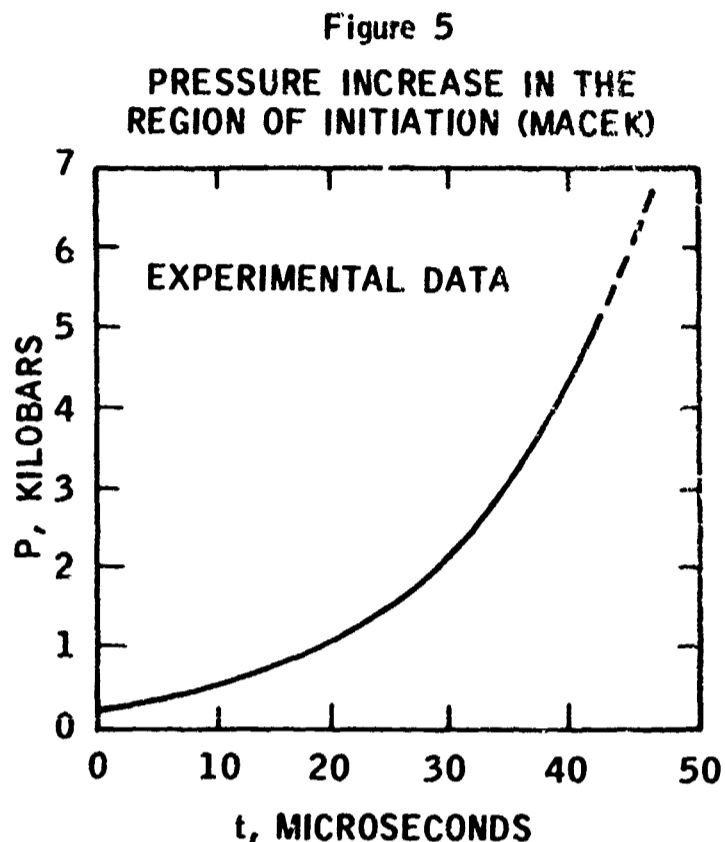
Deflagration - Detonation Transition

The transition from deflagration to detonation in a reacting system is the transition from a burning mechanism propagated by transport phenomena to a burning mechanism propagated by shock waves. The problems of spontaneous transition in gaseous systems has been discussed in the literature since 1903 (160). It has been experimentally determined that, in an accelerating, gaseous deflagration, a shock front runs ahead of the flame; and Kistiakowsky (161) has suggested that transition to detonation in condensed phase materials is also caused by a precursor shock front, i.e., the precursor forms as a result of rapid pressure increase behind the deflagration front and then propagates through the front into the unreacted material. Macek (162) had divided the sequence of events in the transition into three steps:

- Rapid pressure increase behind the burning front, which sends compression waves into the unburnt explosive.
- Coalescence of compression waves in the unburnt explosive, forming a shock wave ahead of the flame.
- Shock initiation of the detonation reaction through a thermal mechanism.

The first two of the proposed steps were studied by Macek.

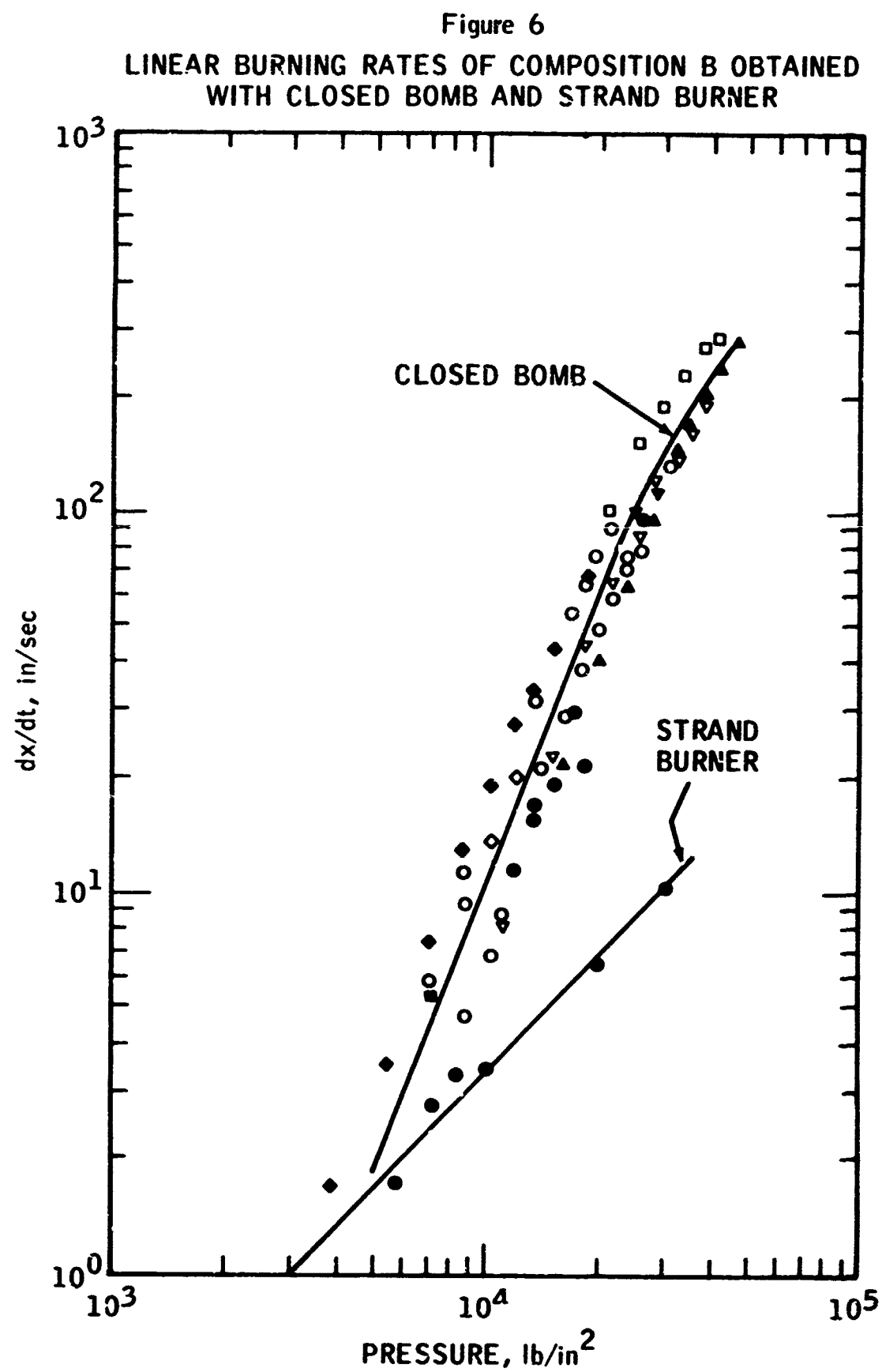
In Macek's experimental work, heavily confined cast cylinders (1.27 cm in diameter, 30 cm long, encased in steel having a wall thickness of 0.625 cm) of two high explosives, DINA and 50/50 pentolite, were initiated thermally by an electrically heated nichrome wire. Propagation velocities were measured over three intervals along the charge, using ionization probes. In those experiments in which the thermally initiated deflagration developed into detonation, the pressure rose from a fraction of a kilobar to the bursting strength of the tube (5-10 kbar) in 40 to 60 μ sec; and it was found that the pressure rise could be adequately approximated using an exponential representation. In an experiment in which the pressure trace was not of the exponential form, indicated in Figure 5, detonation did not occur.



After Macek (162)

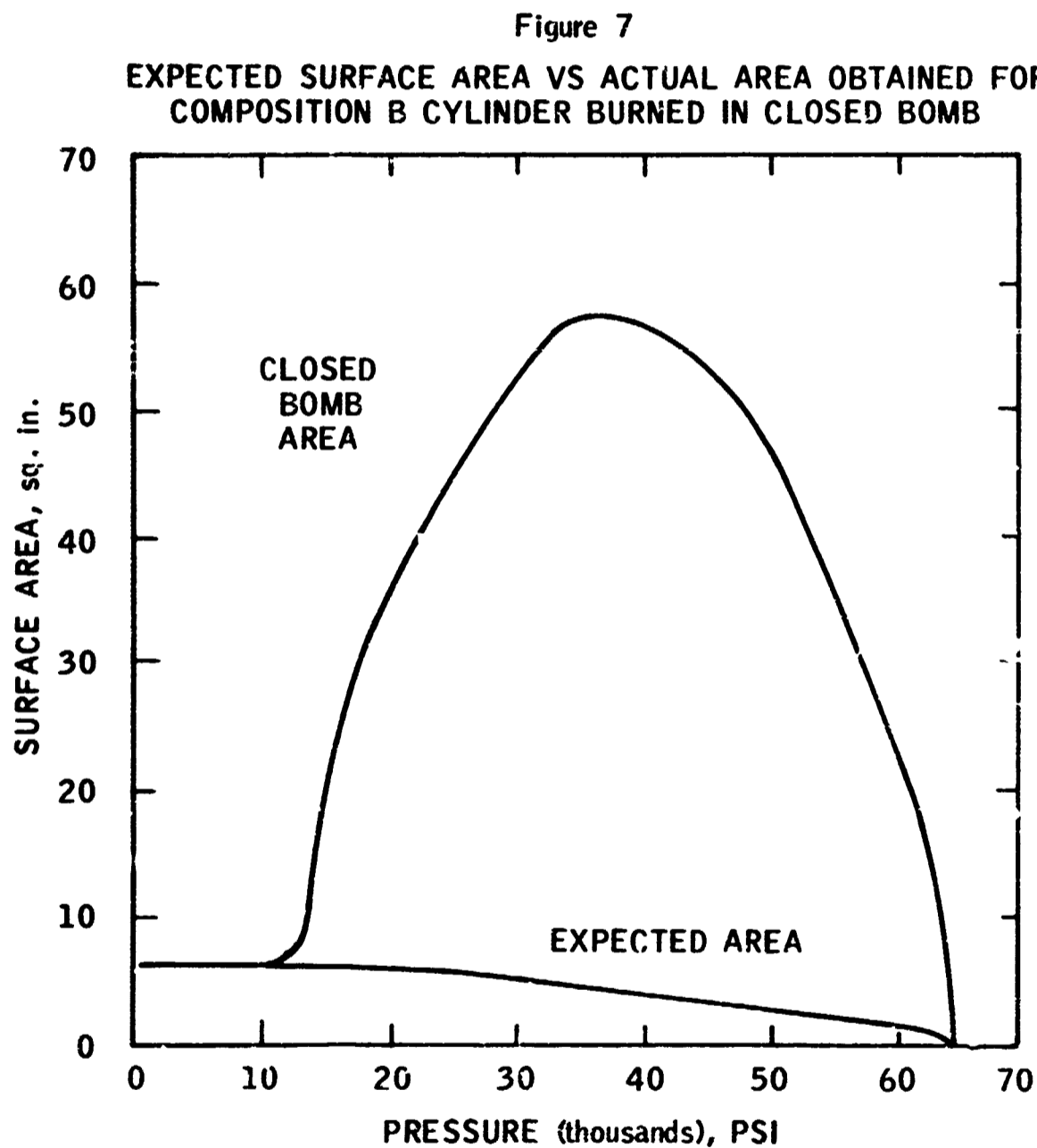
In the mathematical treatment of compression wave coalescence, Macek assumed that a plane of deflagration separated the product gas from the unburnt solid, and that the burning rate was a function of pressure. Compression of the solid was assumed to follow the Tait equation. Using the measured rate of pressure increase, a Riemann analysis was completed, establishing the $u + c$ characteristics and the boundary path between solid and product gas. The compression waves were found to coalesce, forming an incipient shock, when the pressure exceeded several kilobars. The point of shock formation was in qualitative agreement with the experimentally determined point of detonation initiation. This study thus appears to confirm the hypothesis that transition from slow burning to detonation in cast explosives is due to a shock which arises in the burning medium. The approximate nature of the equation of state, and the unknown degree of confinement provided by the expanding tube walls prior to rupture, preclude the possibility of quantitative determination of the point of shock wave formation.

Wachtell and McKnight (163) proposed a method for determination of the detonability of propellants and explosives through a study of deflagration-detonation transition. The basic idea is the concept, first proposed by Kistiakowsky (162) that transition to detonation is essentially a physical process in which the linear burning rate of a bed of material increases to a very high rate, although the individual particles are consumed at a relatively low rate; and that the combustion is shock initiated. It was proposed that a mechanism of this type could be operable with homogeneous as well as heterogeneous materials as a result of surface cracking or crazing occurring under the pressure and thermal stresses accompanying reaction. Wachtell and McKnight performed experiments in which cylinders of TNT, Composition B, and ARP propellant were carefully prepared by machining from specially cast void-free blocks. These samples were burned in closed bombs, using squib initiation. The experimental data indicated a marked deviation from normal burning rate at 6000-8000 psi pressure; Figure 6 presents linear burning rates for Composition B calculated from these data, as well as strand burning rates obtained at pressures from 1000 to 20,000 psi. The burning rate curve is seen to turn sharply upward at about 6000 psi.



After Wachtell and McKnight (163)

Figure 7 indicates the surface area that would be necessary to burn the sample in the bomb at the rate determined in the strand burner: a twenty-fold increase in surface area is indicated. Similar results were obtained for other materials.



After Wachtell and McKnight (163)

Griffiths and Grocock (164) investigated the mechanism by which heavily confined columns of solid granular explosives, e.g., RDX, HMX, PETN, and tetryl, burn to detonation. The experimental work employed cylindrical brass tubes 2.1 inches in external diameter and 0.1 inch in internal diameter. The granulated explosives were placed in the tubes in small increments to avoid density variations, and the prepared samples were initiated by the flame from a lead styphnate ignitor. The progress of the reaction wave was followed with ionization probes, and photographic data were obtained using steel tubes with plexiglas windows.

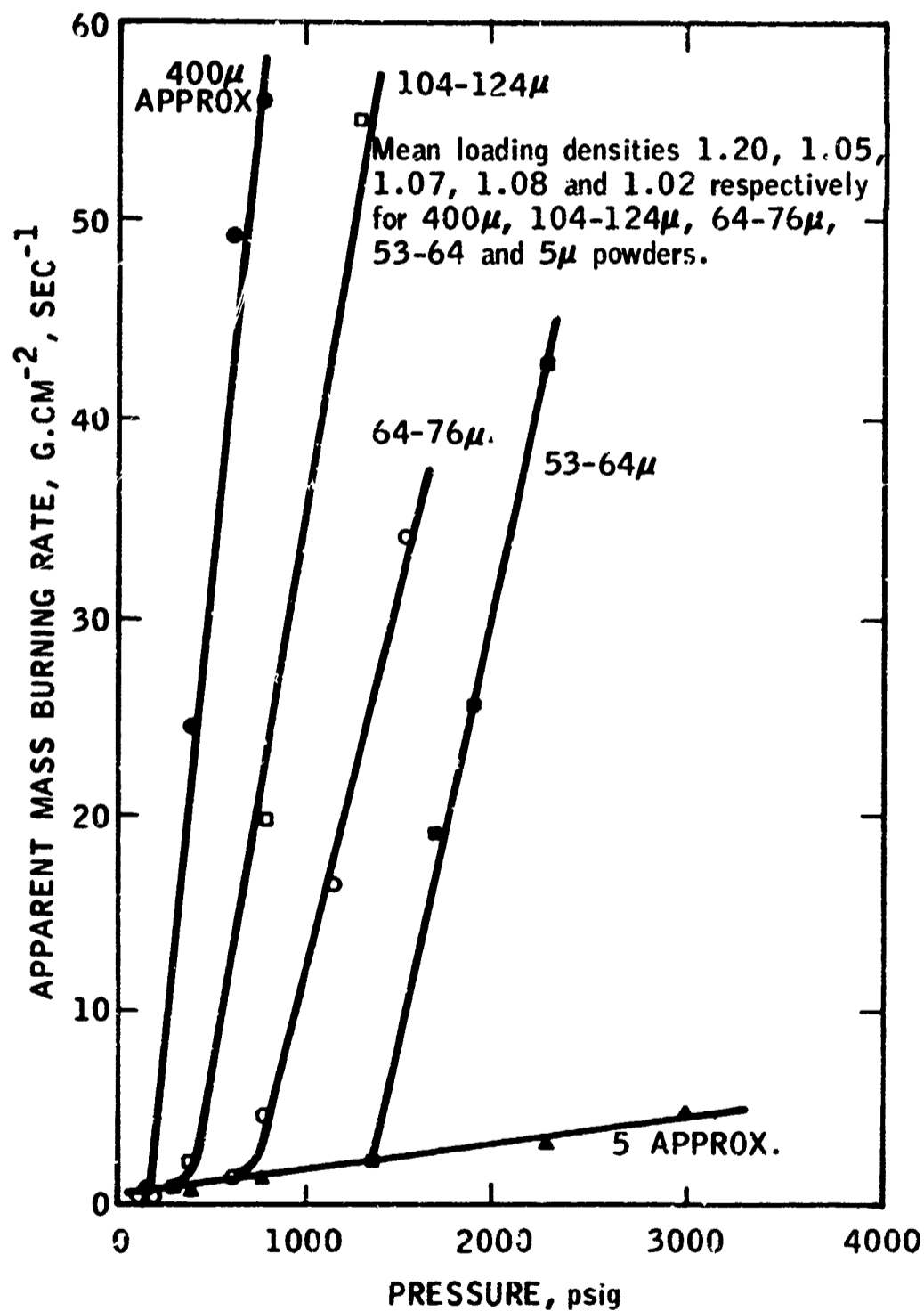
The results showed very low burning velocities in the early stages (1 cm/sec.), accelerating to velocities as high as 2000 cm/sec., and ultimately transition to detonation. The authors break the events into a number of stages.

- A combustion wave, in which the mechanism of heat transfer is conduction, having a propagation rate less than 1 cm/sec.
- A combustion wave, in which the mechanism of heat transfer is convection, having a propagation rate up to 2000 cm/sec.
- Formation of a shock wave from coalescence of compression waves, and initiation of the detonation reaction.

The second, or convective, stage of the transition is the most important, and is dependent upon the temperature of the gas products, the resistance of the explosive to penetration by the gas products, and the energy transfer rates between the gaseous products and the unreacted explosive. The photographic evidence verified the postulated steps for the transition phenomenon.

In a study somewhat similar to that of Griffiths and Grocock, Taylor (165) measured burning rates of PETN, RDX, and HMX powders in a strand burner at elevated pressure. The explosive powders were lightly pressed into paper and Plexiglas tubes having a 6 mm bore diameter, and initiated electrically. The burning rates were determined photographically. Results for HMX are presented in Figure 8.

Figure 8
BURNING RATES OF PRESSED HMX POWDER AT VARIOUS PRESSURES



After Taylor (165)

These results indicate that, for a given powder particle size, a transition pressure exists, above which the burning rates are considerably higher than normal for such materials. It was postulated that the rapid burning rates were due to pressure gradients which forced the combustion gases into the porous powder, enabling convective energy transfer ahead of the burning front. The photographic evidence supported this postulate. The transition pressures occur as a result of a layer of molten explosive which forms above the powder. At low pressures, the combustion gases are unable to penetrate the liquid layer and enter the porous explosive; the transition pressure represents the point where the liquid layer is first penetrated due to the pressure gradient.

The convective burning model thus provides a mechanism for the pressure to build to the point where a shock wave can be formed which is capable of initiating detonation. In principle, the high convective burning rates could be stabilized short of detonation by establishing a stable pressure gradient, but this would be possible only with porous materials; and transition to detonation is the usual result. Andreev (166) has fully discussed the convective mechanism for deflagration acceleration in porous materials. He considers the major factors influencing the transition to be:

- High gas permeability of the granular charge.
- High temperature combustion products.
- High burning rate.
- High "ignitability" for the explosive.

He notes that the transition to detonation is always accompanied by an extension of the zone of combustion such that the deflagration reaction is occurring over a large area. He attributes transition in liquid systems to the formation of turbulence within the zone of reaction.

The most recent study of deflagration-detonation transition was carried out at the Naval Ordnance Laboratory (167). This work involved Macek (162) type experiments under improved conditions. Cased pentolite in seamless steel tubes was employed, and the reaction wave propagation was followed with strain gages, ionization probes, and a framing camera. Initiation was by burning squib. This work demonstrated a low velocity (subsonic) ionization front which was pressure initiated and pressure propagated. This was definitely not a flame front propagated by transport mechanisms. The critical initiation pressure for the observed ionization phenomena was of the order of two kilobars. The ionization front was observed to occur in all experimental shots, even those which did not grow to detonation; thus, it appears that the ionization front is not related to the transition phenomena.

It was determined that two criteria had to be met in order for the deflagration reaction to grow to detonation:

- The explosive must burn rapidly enough so that a high pressure is reached in a confined area.
- The explosive must be sufficiently shock sensitive to be initiated by this pressure.

These studies, taken together and in conjunction with additional numerical studies (168,169), provide conclusive evidence that the hypothesis presented concerning the events leading to the transition of burning to detonation is in fact a reasonable one. The data indicate the presence of pseudostable convective burning rates over relatively long time periods during the transition process.

The two preceding discussions of "shock-initiation of detonation" and "deflagration-detonation transition" demonstrate that considerable advances have been made in understanding the mechanism of initiation of explosives by a variety of stimuli. Many systems can be initiated by stimuli which are below those necessary for direct detonation initiation; but such initiations do not always cause detonation. The growth of reaction is dependant on geometry and other properties of the material. Heterogeneous explosives are more difficult to understand from the initiation viewpoint, and the possibility of extending advances in the understanding of homogeneous initiation to the heterogeneous case is not probable. Much more work is required to establish a good basis for understanding heterogeneous initiation.

APPENDIX

NOMENCLATURE

Symbols

a	=	Length of reaction zone (L)
c	=	Sound velocity (Lt^{-1})
D	=	Velocity of wave with respect to observer (Lt^{-1})
d	=	Charge diameter (L)
d_{eff}	=	Effective charge diameter (L)
E	=	Specific Internal Energy (L^2t^{-2})
η	=	Eugoniot function (L^2t^{-2})
h	=	Length of "critical" region (L) or Planck's Constant (mL^2t^{-1})
k	=	Boltzmann's constant ($mL^2t^{-2}e^{-1}$)
L	=	Charge length (L)
L_m	=	Maximum effective charge length (L)
M	=	Mass rate of flow ($mL^{-2}t^{-1}$)
P	=	Pressure ($mL^{-1}t^{-2}$)
Q	=	Chemical reaction energy (L^2t^{-2})
Q_r	=	Energy released by a wave (L^2t^{-2})
Q_w	=	Energy required to support a wave (L^2t^{-2})
R	=	Charge radius (L)
R_o	=	Gas Constant ($mL^2t^{-2}e^{-1}$)
r	=	Radial coordinate or charge radius (L)
T	=	Temperature (θ)
u	=	Material velocity (Lt^{-1})
V	=	Specific volume (L^3m^{-1})
v	=	$u-D$ (Lt^{-1})
ΔH	=	Activation energy per mole (mL^2t^{-2})
ΔS	=	Activation entropy per mole ($mL^2t^{-2}e^{-1}$)
α	=	Covolume ($m^{-1}L^3$)
ρ	=	Density (mL^{-3})
τ	=	Reaction time (t)

Subscripts

o	=	Refers to initial state
l	=	Refers to final state
c_j	=	Refers to Chapman-Jouguet conditions
i	=	Refers to ideal conditions

Superscripts

o	=	State ahead of shock
l	=	State behind shock

Figure 1
HUGONIOT CURVE

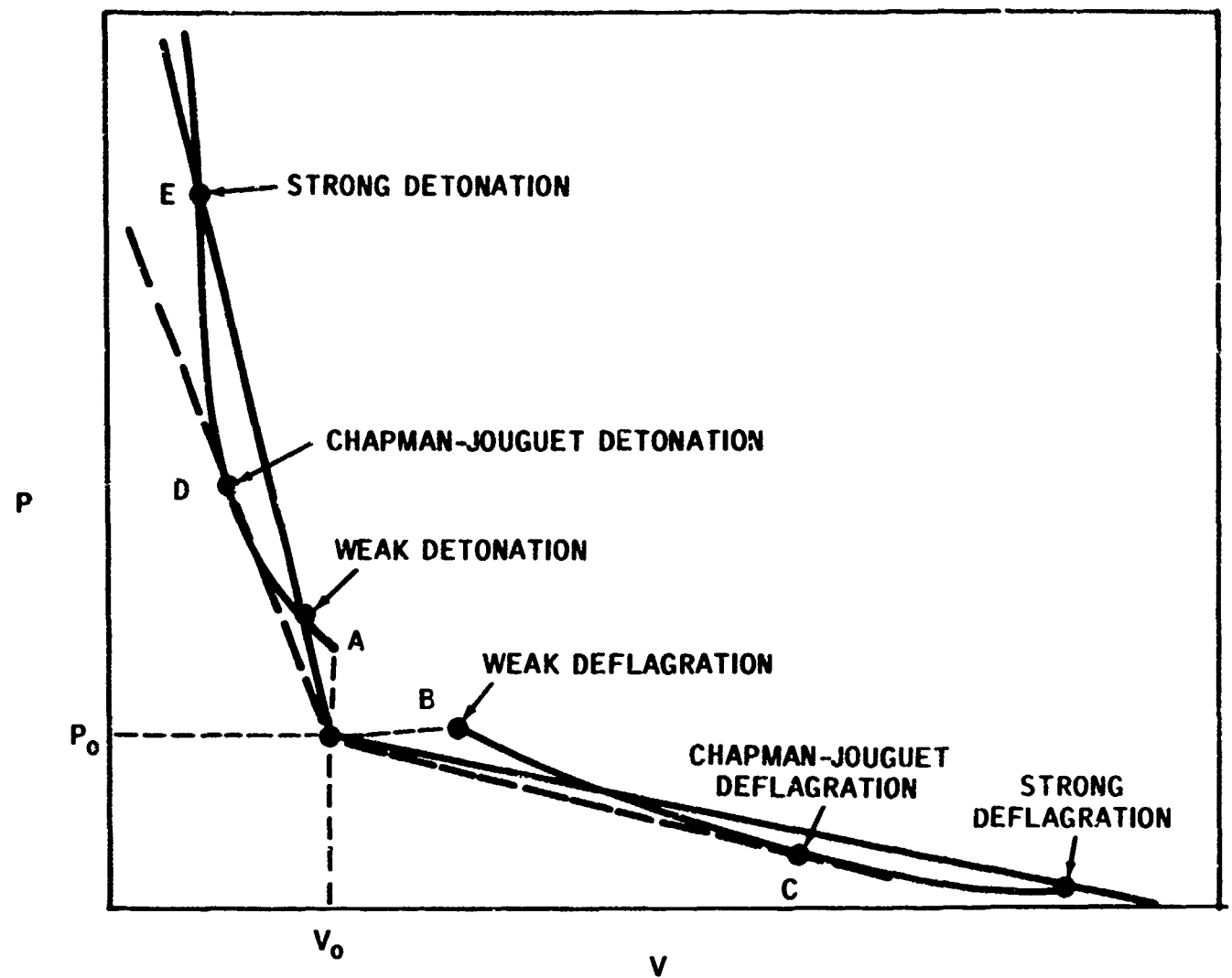


Figure 2
DETONATION HUGONIOT

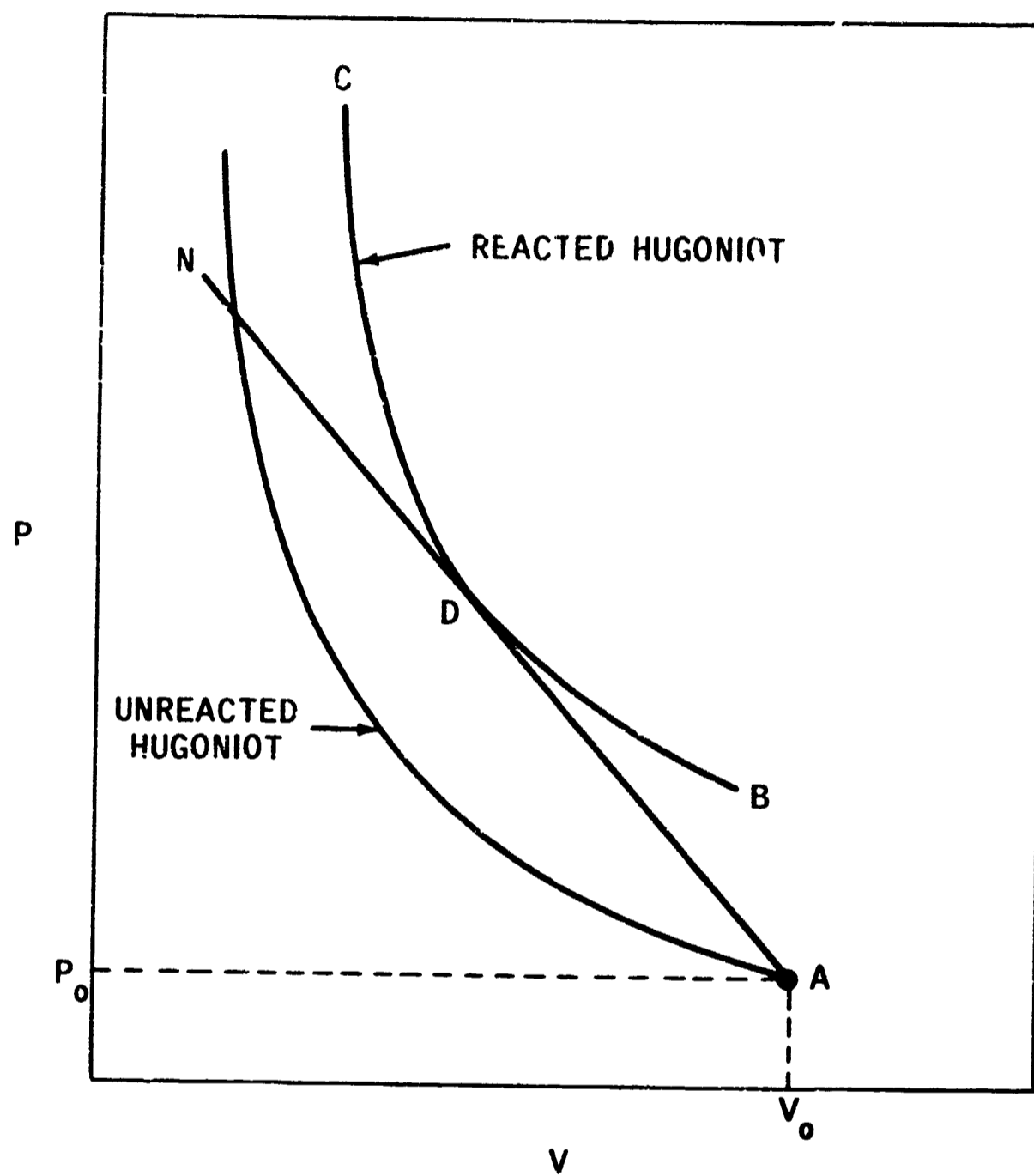
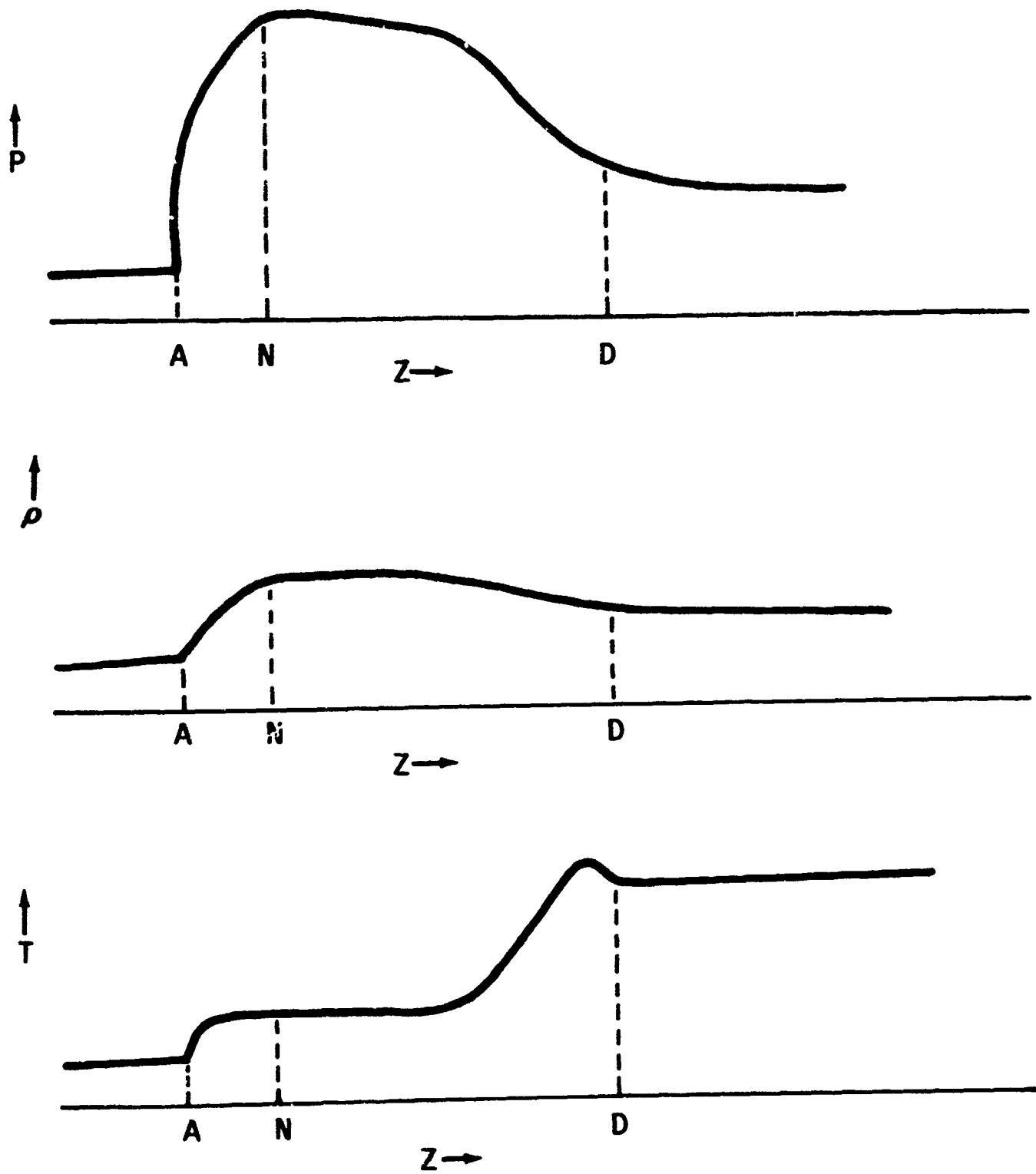


Figure 3
PRESSURE, DENSITY AND TEMPERATURE
THROUGH THE REACTION ZONE



After Hirschfelder, et.al. (205)

PRECEDING
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2.3. Phenomena Intermediate Between Deflagration and Detonation

Current theories do not admit the possibility of stable reaction regimes having propagation rates and pressures intermediate between those of deflagration and detonation. There exists, however, a growing body of experimental evidence of reaction waves which appear to fall into the intermediate area under certain conditions of confinement and initiation. Most of these are probably cases of familiar phenomena under limiting and not-very-practical conditions, but one or two may be new and useful phenomena.

2.3.1. Phenomena Which Have Been Observed

Observations of condensed-phase reaction waves possessing velocities which appear to be intermediate between conventional deflagration and conventional detonation can be grouped into four categories:

- Ideal detonation in low-density media
- Nonideal detonation
- LVD in liquids and solids
- Ultra-fast (probably convective) deflagration

Each of those categories is discussed below. Reactions in the process of transiting from deflagration to detonation are excluded from consideration because their intermediate velocity is by definition only transient.

Ideal Detonations in Low-Density Media

In so-called "ideal detonations," all of the detonation properties can be calculated, using the thermohydrodynamic theory and an appropriate equation of state for the detonation products. "Ideal" means that all the environmental parameters, such as charge diameter, initiation strength, etc., are above the critical level; so that the detonation processes are determined by only two independent parameters, the initial chemical composition and the loading density of the explosive material. Thus, the detonation velocity of a given chemical composition is a function only of its initial density. Experimental studies of ideal detonations have yielded three different types of relationships between detonation velocity and loading density.

For most condensed phase explosives, the relation between loading density and detonation velocity is linear, and may be expressed by the equation

$$D = D_{\rho_1} + n(\rho_0 - \rho_1)$$

where D = the detonation velocity at the postulated loading density

D_{ρ_1} = the known detonation velocity at a known loading density, ρ_1

n = the slope of the velocity/density curve

ρ_0 = the postulated loading density

ρ_1 = the known loading density

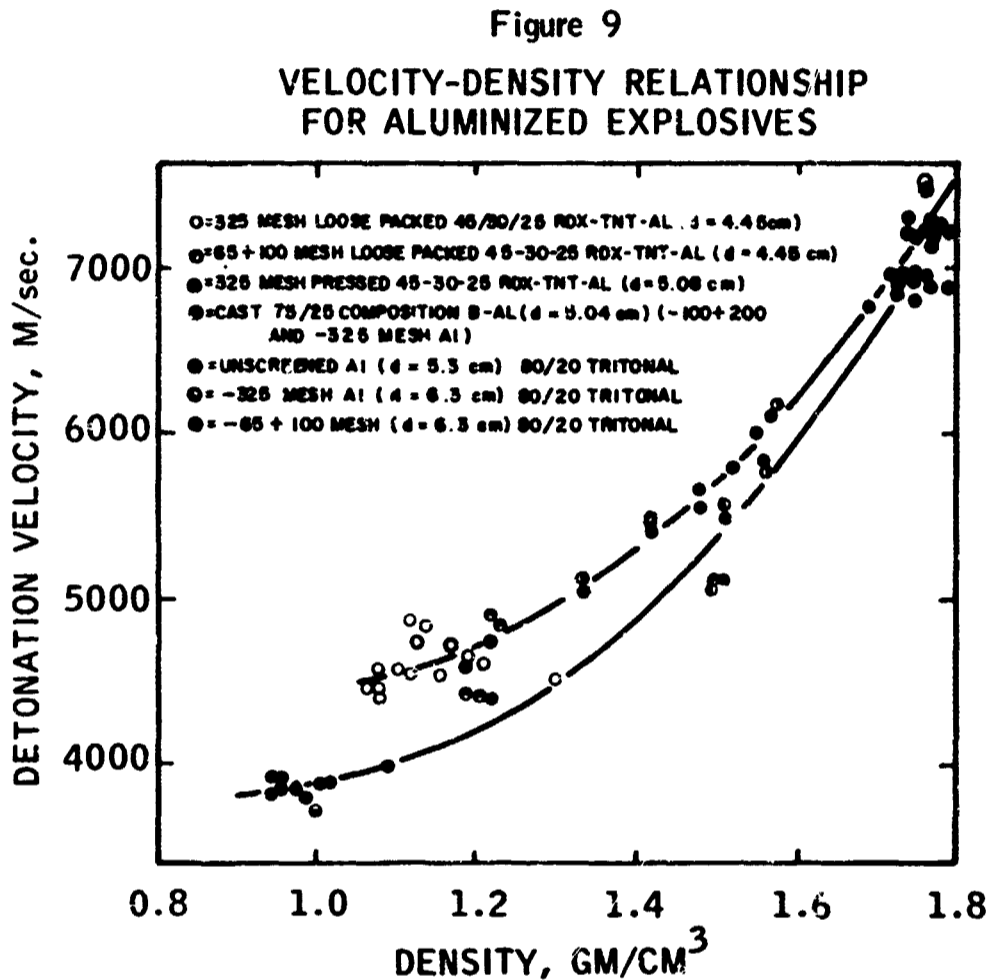
The following table lists equation parameters for most important explosive materials (53).

TABLE 1

IDEAL DETONATION VELOCITY DATA

<u>Explosive</u>	<u>Density</u> <u>1</u>	<u>D</u> <u>1</u>	<u>M</u>
TNT	1.0	5,010	3,225
PETN	1.0	5,500	3,950
50/50 Pentolite	1.0	5,480	3,100
RDX	1.0	6,080	3,590
Composition A	1.6	8,180	4,000
Composition B	1.6	7,540	3,080
Composition C	1.5	8,100	--
Composition C-2	1.57	7,850	--
Tetryl	1.0	5,600	3,225
65/35 Tetrytol	1.6	7,300	3,400
Picric Acid	1.0	5,255	3,045
EDNA	1.0	5,910	3,275
Ammonium Picrate	1.0	4,990	3,435
Nitroguanidine	1.0	5,460	4,015
Lead Azide	4.0	5,100	560
Mercury Fulminate	4.0	5,050	890
50/50 Amatol	1.0	5,100	4,150
50/50 Sodatol	1.0	4,100	2,580
DINA	1.0	5,950	2,930
NENO	1.0	5,530	3,680
Fivonite	1.0	5,215	3,410
Sixonite	1.0	5,670	3,360
NG	1.59	7,800	--
60/40 NG-DNT	1.50	7,000	--
60/40 EDNATOL	1.60	7,510	3,325

Exceptions to the linear density-velocity relationship for condensed phase materia : generally involve explosive compositions containing aluminum. The following figure presents experimental data for typical aluminized explosive compositions.



After Anderson and Pesante (170)

The reason for the observed nonlinearity in such curves stems from the fact that the heat of explosion (which affects the detonation velocity) is largely determined by the ratio Al_2O/Al_2O_3 in the reaction products, and thermodynamics dictates that this ratio is large at low loading densities and small at high loading densities. The formation of Al_2O is endothermic with respect to the overall reaction, while the formation of Al_2O_3 is quite exothermic. Thus, the heat of explosion is largely determined by the ratio Al_2O/Al_2O_3 in the reaction products; and the apparent heat of explosion is effectively a function of loading density.

Aluminized explosives have long been noted for their low brisance and high blast potential. This phenomenon can be explained via considerations similar to those used above. A great amount of the reaction energy is released too late to affect the detonation wave but sufficiently soon to contribute to the destructive or blast potential. For this reason, such materials are ideally suited to blasting operations.

The third relationship between detonation velocity and loading density applies to gaseous detonations. In such cases, the detonation velocity has been found to be a slowly varying function of the loading density. This is due to the influence of density on molecular weight and its effect on particle velocity. Theoretical computations for gaseous detonations have been verified by experimental data both with regard to the absolute magnitude of the detonation velocity and the slope of the velocity-density curve (171).

Regardless of the form of the loading density-detonation velocity relationship, it is apparent that low detonation velocities can be obtained by employing low loading densities. The lower limit for such phenomena is sonic velocity in the unreacted material, since by definition detonation is a shock initiated phenomenon. Sonic velocities for condensed phase materials usually range from 1500 to 3000 meters per second. Thus, experimental examples of reaction wave phenomena which appear to be intermediate between conventional detonation and conventional deflagration can be found which are in fact detonations under conditions of low loading density.

An example of detonation at low loading density is the use of nitroguanidine for explosive welding by Sadwin (172). This material is reported to be detonable at bulk densities from 0.11 to 1.7 gm/cm³, corresponding to computed detonation velocities from 1890 to 8270 m/sec and detonation pressures as low as one kilobar. Sadwin gives the relationship between detonation velocity and bulk density as

$$D = 1445 + 4015\rho$$

and that between detonation pressure and bulk density as

$$P = 77\rho^{1.94}$$

where D = detonation velocity in meters/second

P = detonation pressure in kilobars

ρ = bulk density in grams/cm³

The density range studied experimentally was from 0.6 to 1.2 gm/cm³, corresponding to a range of 28 to 109 kbars in detonation pressure and 3850 to 6250 m/sec in detonation velocity.

In a somewhat different aspect of low density experiments, Abegg and co-workers (173) have evaluated a series of plastic/explosive compositions.

They found that detonation pressures as low as 25 kilobars could be obtained from 10% PETN/90% nitropolyurethane, and as low as 22 kilobars from 70% PbN₆/30% polyurethane. The pressures followed the relationship:

$$P = 0.10 \rho \frac{D^2}{4}$$

where P = detonation pressure in bars

ρ = density in grams/cm³

D = detonation velocity in m/sec.

Observed detonation velocities ranged from 3000 to 7000 m/sec., with most results lying between 3500-6000 m/sec.

Abegg, et al., (174) have also investigated a series of high-density coordination compounds of the type $\text{Cu}(\text{tmen})_2(\text{IO}_3)_2$ which yielded detonation pressures as low as 11 kilobars and as high as 230 kilobars ("tmen" = tri-methylenediamine). Oxygen balance and consolidation density were important parameters. Heavy ions lowered the detonation velocity but the increased density of the total molecule tended to keep the detonation pressure high.

The current state of the art is such that the use of low loading densities or plastic/explosive mixtures enables one to obtain detonation velocities as low as 1500 m/sec. and detonation pressures as low as one kilobar. The work of Abegg, et al., (173,174) enables the selection of detonation pressure from 20-90 kilobars in 10 kilobar steps by appropriately controlling the properties of the explosive. It is anticipated that work in progress will enable similar control to be established for pressures from 1 to 10 kilobars in 1 kilobar steps. These capabilities provide an extremely convenient and useful laboratory tool.

Nonideal Detonation

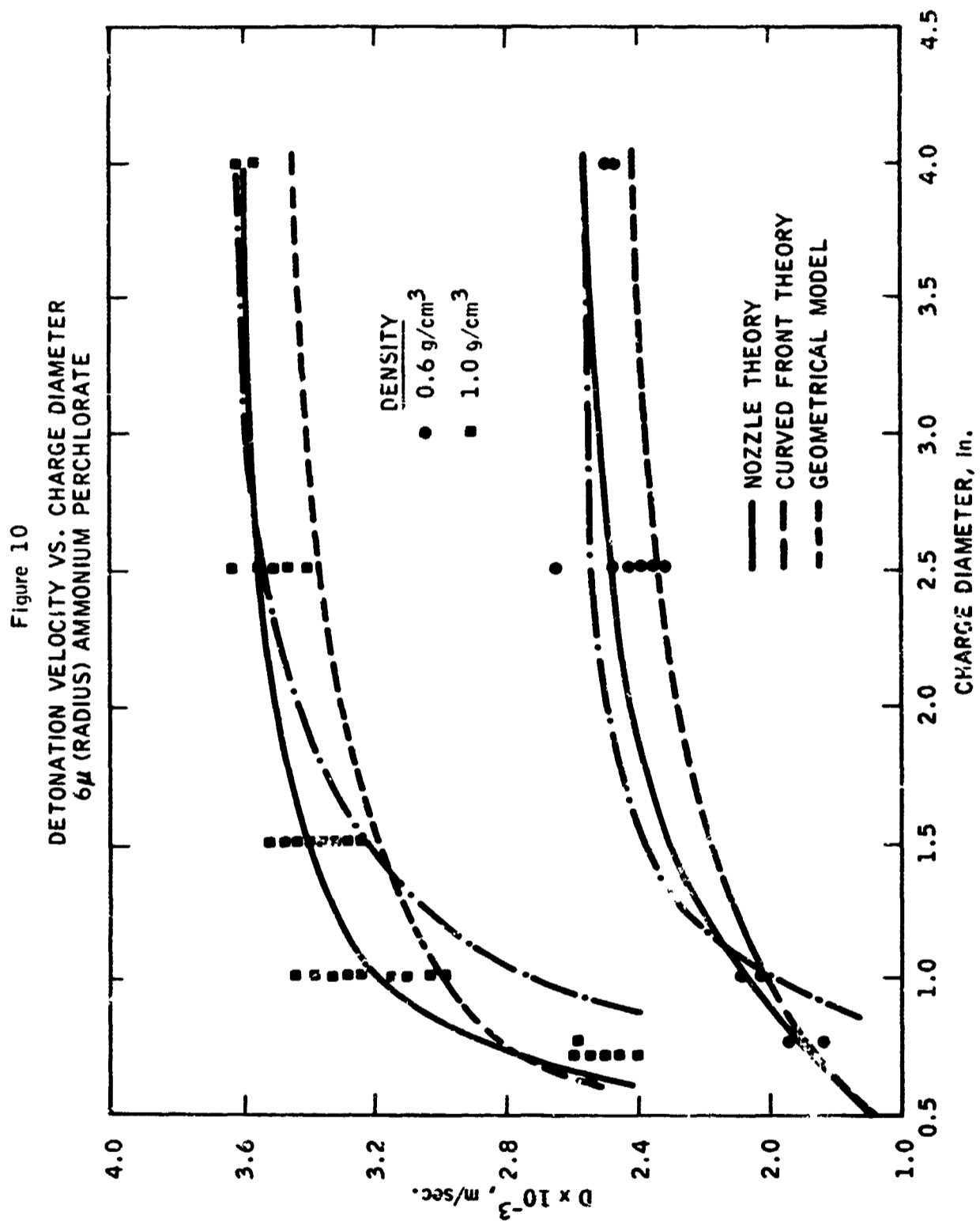
It has long been observed that detonations propagating in charges of small diameters do so at less than the theoretical detonation velocity. If the charge diameter be made sufficiently small, the detonation wave fails to propagate. At diameters larger than the failure diameter, the detonation velocity increases with increasing charge diameter, asymptotically approaching the theoretical value. These facts give rise to the definition of two important parameters for a given explosive composition and physical state: (1) the critical diameter, d_c , below which wave propagation fails, and (2) the minimum diameter for ideal detonation, d^* , above which the detonation velocity attains its maximum theoretical value. Detonations propagating in charges having diameters between d_c and d^* are termed "nonideal."

The extent of the region of nonideal detonation is dependent upon the rate of reaction of the explosive and the degree of confinement provided by the surroundings. Both d_c and d^* are increased by increasing particle size, while both quantities are reduced by increased confinement. Therefore, for a given diameter, detonation velocity is increased by increased confinement.

Nonideal detonation is generally associated with the presence of a finite reaction-zone length or, equivalently, a finite reaction time (51,53,170,175,176). Since the reaction zone length is appreciable, the high pressure within the zone has time to produce appreciable lateral expansion before the reaction reaches completion. Thus, a fraction of the energy produced by the reaction is dissipated from the sides of the charge; and that fraction is not available to maintain the detonation. Mathematical analysis (175) suggests that the effect of lateral losses on the detonation velocity becomes significant when the radius of the charge is of the same order of magnitude as the length of the reaction zone. The energy losses also give rise in many cases to incompleteness of reaction and consequent lowered energy yields.

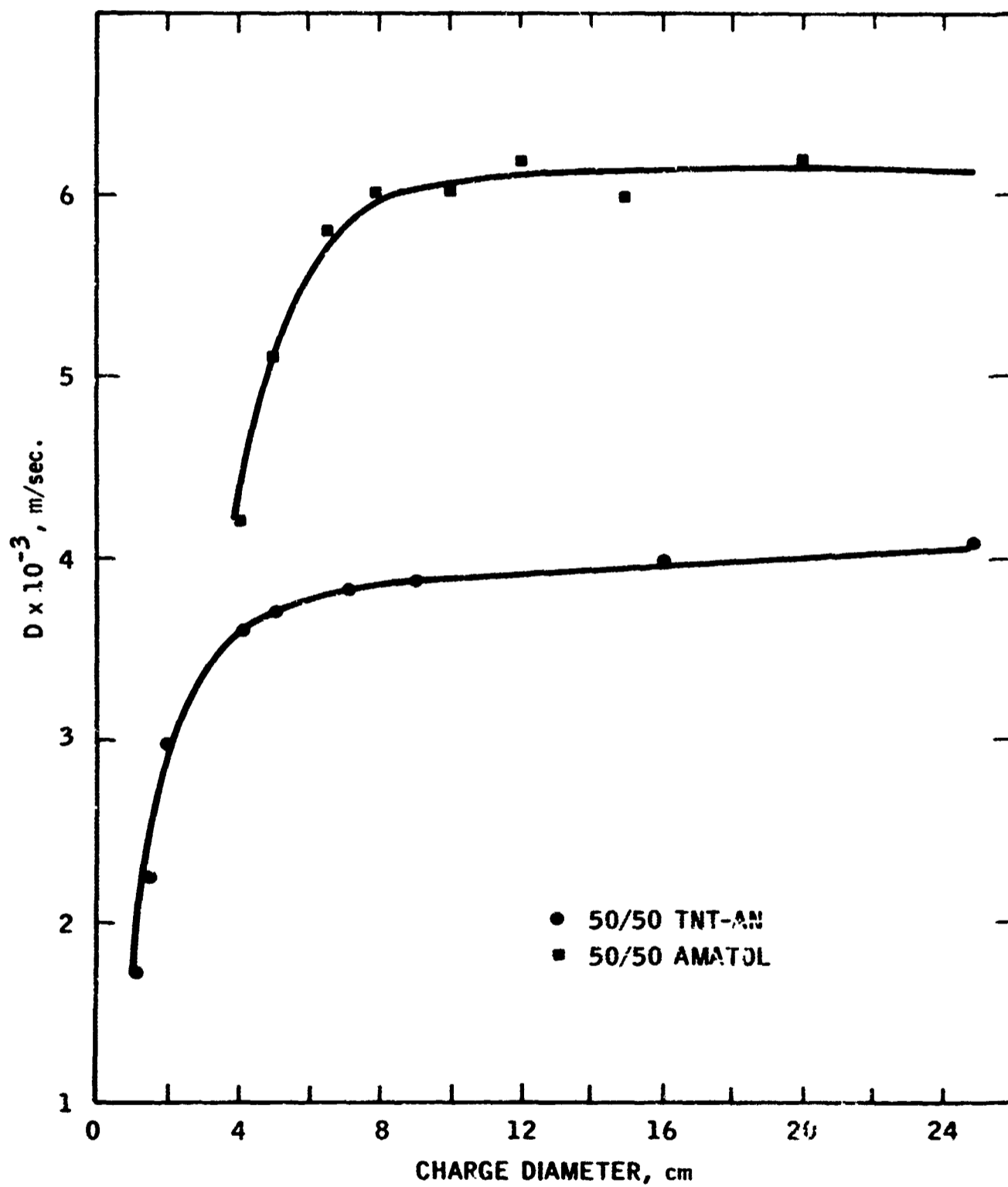
Figures 10 and 11 (on pages 49 and 50, following) present experimental data for detonation velocity as a function of charge diameter for several explosive compositions (170,53). One can observe that very low detonation velocities can be obtained at charge diameters near the critical diameter. It should be emphasized that these propagation velocities are stable, steady, and predictable for any particular condition of charge diameter and confinement. Once again, the lower limit for such phenomena is the sonic velocity of the unreacted material. Thus, proper selection of charge diameter and confinement for a given explosive composition can yield a detonation velocity which appears to fall between the limits of conventional detonation and conventional deflagration.

The use of charge diameters near the critical diameter for a given explosive composition can therefore yield controlled detonation velocities as low as about 1500 m/sec. Detonation pressures, however, cannot be controlled to the same degree as offered by the use of the plastic/explosive mixtures previously discussed.



After Cook (53)

Figure 11
DETONATION VELOCITY VS. CHARGE DIAMETER



After Cook (53)

LVD in Liquids and Solids

For many years, experimental evidence has existed suggesting that certain explosives can detonate at two different rates. The "normal" or "high velocity" detonation rate corresponds to that predicted by current thermohydrodynamic theory, while the "low velocity" detonation propagates at about sonic velocity for the unreacted material. Both rate regimes appear to be stable, steady-state, reaction wave phenomena. The term "LVD" is a jargon term which has come to mean a low velocity detonation in a material which can also undergo a normal, high velocity detonation.

Liquid Explosives

The earliest observations of dual propagation rates were in nitroglycerin. Stettbacher (177), as early as 1919, noted two propagation rates for nitroglycerin, and suggested that the two known crystalline forms of solid nitroglycerin were responsible for the two detonation regimes observed. Other investigators (178) supported this theory, even though it was inconsistent with the experimental observation that both high and low velocity regimes could be initiated in either system by using primers of different severity.

Clapham (179) investigated the effect of charge diameter and severity of initiation on the detonation velocity of nitroglycerin. His results are summarized in Table 2.

TABLE 2

<u>Charge Diameter, in.</u>	<u>Detonation Velocity, m/sec</u>			
	<u>#2 Fulminate</u>	<u>#6 Fulminate</u>	<u>#8 Fulminate</u>	<u>#8 Briska</u>
1/4	850 910	810 890 1,030	1,350	8,130
1/2	2,530	1,940 2,090	1,780	8,700
3/4	2,130	1,970 2,030	1,750	8,250 8,390
1	2,190	2,020 2,030	---	8,130
1 1/4	1,740	1,780 2,010	---	8,140
1 1/2	---	1,910	---	---

These results indicate that the detonation velocity is independent of charge diameter when a strong initiator is used. Less severe initiation results in "low velocity" detonation, which increases in velocity with charge diameter as charge diameter is increased from one-quarter to one-half inch. For larger charge diameters, the low velocity detonation propagates irregularly. Ratner (180,181) measured "low velocity" propagation rates in nitroglycerin and other organic nitrate systems with similar results.

Joyner (179) measured reaction wave propagation velocities in mixtures of hydrazoic acid and water. His results are summarized in Table 3.

TABLE 3

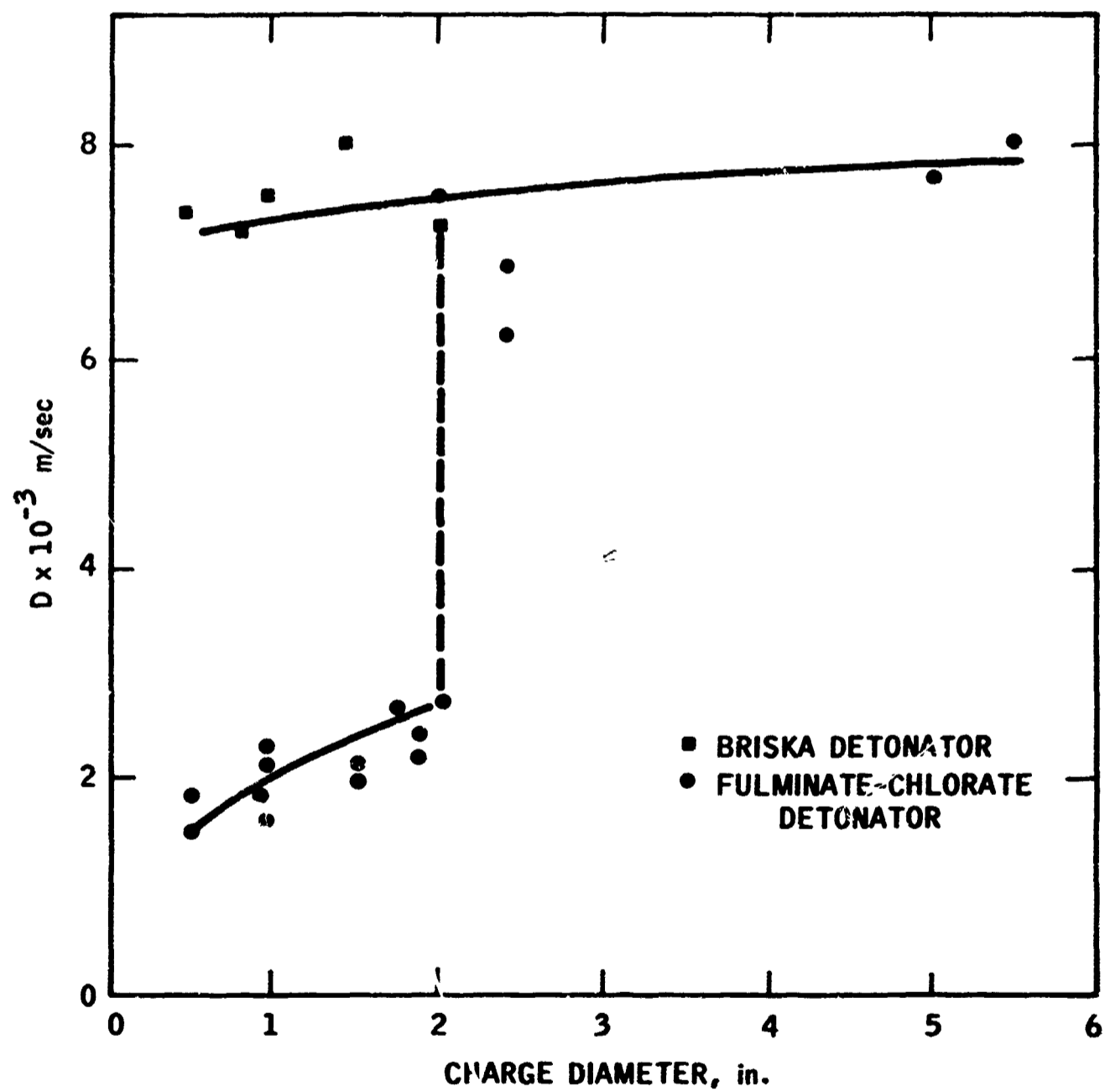
PROPAGATION OF EXPLOSION IN HYDRAZOIC ACID-WATER SOLUTIONS

<u>HN₃ Conc.</u> <u>g/100 g Solution</u>	<u>Normality</u>	<u>Velocity, m/sec</u>
80	22.1	8,500
70	18.0	7,300
67	17.2	7,100
65	16.8	1,700
61	15.7	630
60	15.4	384
54	13.8	233
48	12.2	40
38	9.8	2.5
30	7.3	1.0
16.7	4.0	0.14

This work provides a striking example of the variation in propagation rates that can be obtained under proper conditions in liquid explosive systems.

Gelatinous explosives are really thickened liquid explosives and hence should be considered in the same category with liquid explosives. Experimental work conducted with blasting gelatine has shown evidence of two wave propagation velocities analogous to liquid nitroglycerin. Figure 12, following, presents the relation between detonation velocity and charge diameter for a typical blasting gelatine (179).

Figure 12
PROPAGATION FAILURE IN BLASTING GELATINE



After Taylor (179)

As in other systems, the severity of initiation has been shown to control the mode of detonation. The high velocity mode is seen to propagate essentially at the theoretical thermohydrodynamic rate of 7500 m/sec., while the low velocity mode propagates at a velocity near sonic for the unreacted gelatine.

It has been experimentally demonstrated that reaction wave propagation phenomena in gelatines are strongly dependent upon aeration. Non-aerated gelatines generally fail to propagate reaction when initiated by weak detonators; while well aerated mixtures, with fine air bubbles, propagate at high velocity. Gelatines with intermediate aeration exhibit low velocity detonations under the same initiation conditions. High velocity detonation is achieved in non-aerated gelatines if a sufficiently powerful initiator is employed (179).

Gurton (182) studied the effects of pressures and included gas bubbles on the propagation of low velocity detonations in nitroglycerin. His experimental results are summarized in Table 4.

TABLE 4

DETONATION OF NITROGLYCERIN, CHARGE DIAMETER 7/16" DIAMETER

<u>Detonator</u>	<u>Pressure, Atm.</u>	<u>Gas</u>	<u>Velocity, m/sec</u>
#1	1.0	Air	1410, 1160, 950
#1	11.3	Methane	No Reading
#1	30.3	Methane	No Reading
#1	71.0	Methane	No Reading
#4	1.0	Air	1250
#4	1.0	Air	7690
#4	14.3	Methane	6670
#4	17.7	Methane	1205
#4	20.5	Methane	1040
#4	32.7	Methane	1010
#4	44.3	Methane	807
#4	67.7	Methane	909

When weak initiation was used, low velocity detonations were observed at one atmosphere pressure. Subjecting the charges to higher initial pressure, and replacing the dissolved air with methane, reduced the detonation pressure to the point where velocity traces were not obtained, although the charges exploded completely. Strong initiation occasionally led to high velocity

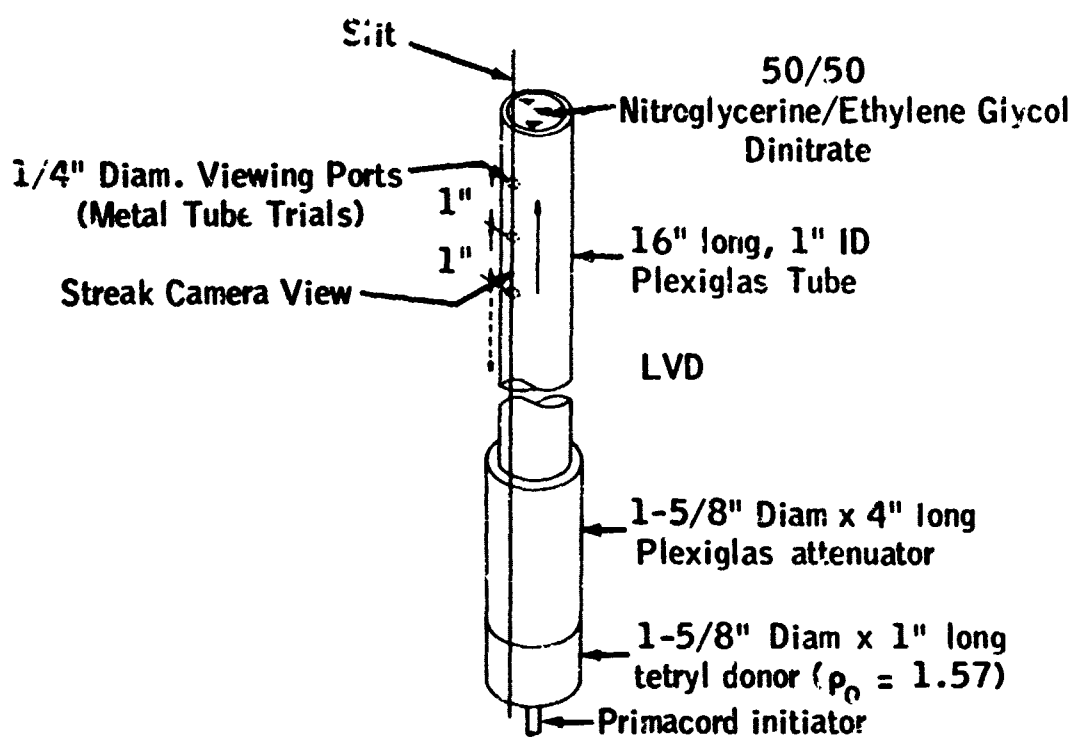
detonation when the included gas was either air or methane. An increase in the external pressure appeared to reduce detonation velocity, but pressures over 70 atmospheres failed to prevent propagation. These observations are what would be expected if the initiation and propagation of low velocity detonation in nitroglycerin is dependent upon the presence of gas bubbles in the liquid. During the course of this work, however, nitroglycerin charges were carefully degassed under high vacuum, placed under external pressure in excess of 60 atmospheres and fired immediately. Low velocity detonations were observed for these cases even though it was very unlikely that any gas bubbles could have remained in the charges.

Nitroglycerin in cylindrical, square, and triangular cross section confinements has been observed to detonate in the low velocity mode (183). Photographic results showed that the slow propagation of the detonation wave could be attributed to a shock wave formed during initiation, propagating in the confining medium at a much higher velocity than in the explosive. The shock wave propagation in front of the detonation wave caused the formation of discontinuities in the charge and the presence of the discontinuities decreased the speed of sound in the nitroglycerin to about 700 m/sec. The low velocity detonation was seen to propagate at 800-1000 m/sec.

In recent years, several accidents have occurred in nitroglycerin and liquid monopropellant manufacturing facilities. These accidents have resulted from relatively mild stimuli and have indicated that the study of low velocity detonations should play an important role in the characterization of the hazards of liquid explosive systems. For this reason, efforts to define more clearly the low velocity regime for liquid explosives have been initiated at the Explosives Research Center of the U.S. Bureau of Mines and at Stanford Research Institute. These efforts have provided the most comprehensive experimental study of low velocity reaction wave phenomena accomplished to date. Three explosive systems have been considered, a 50/50 mixture of nitroglycerin and ethylene glycol dinitrate, mixtures of nitro-methane-tetranitromethane, and Cavea B 110, a mixture of nitric acid and fuel.

The experimental charge arrangement used in the most recent Explosive Research Center work (184) is presented in Figure 13.

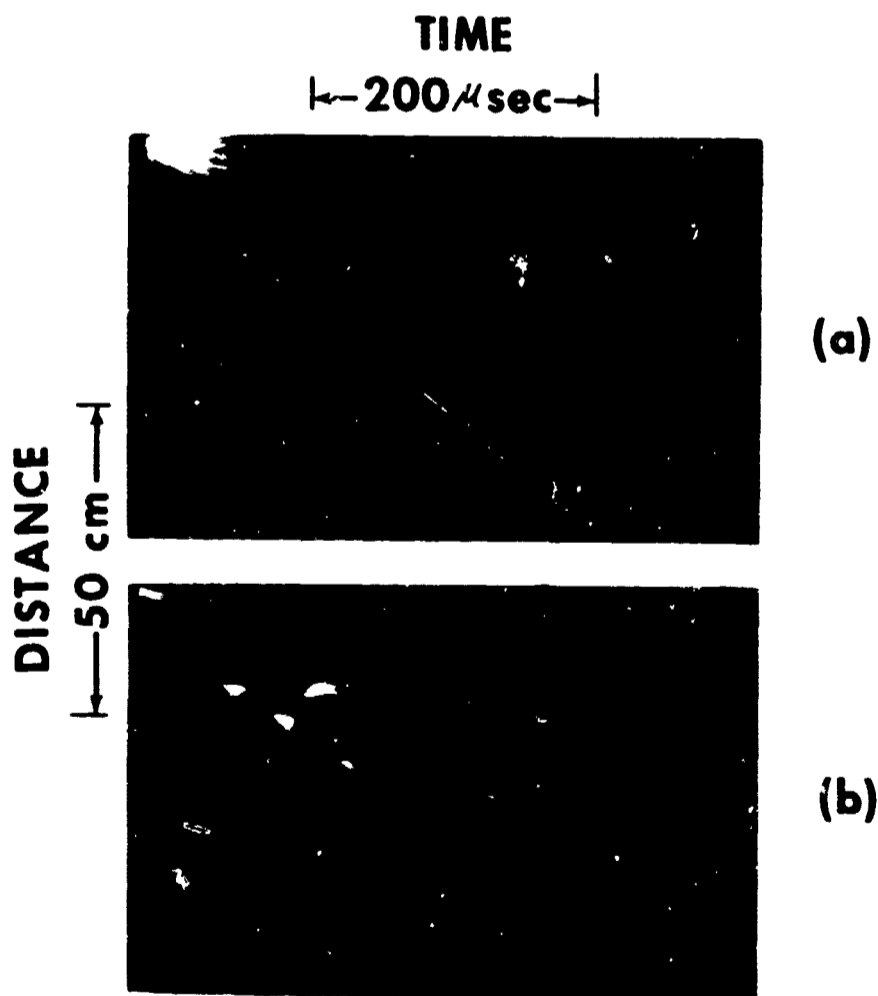
Figure 13
BU MINES EXPERIMENTAL APPARATUS



After Watson, et.al. (184)

A tetryl charge coupled to a Plexiglas shock attenuator serves as a shock donor, and the donor-attenuator combination delivers a peak pressure of approximately 2.4 kilobars. This pressure level has proven adequate to initiate low velocity reaction wave phenomena in nitroglycerin-ethylene glycol dinitrate systems. The receiver consists of a cylindrical tube sixteen inches long and one inch in diameter. Experiments have been performed using containers constructed of various materials having a range of wall thicknesses. The stability of the observed reaction wave phenomena was determined by continuous streak camera records of the progress of the wave over the length of the charge. Typical streak camera records illustrating both stable and unstable low velocity detonations are presented in Figure 14.

FIGURE 14
TYPICAL STREAK CAMERA RECORDS ILLUSTRATING:
(a) STABLE LOW-VELOCITY DETONATION IN A
1/4-INCH WALL PLEXIGLAS TUBE; (b) UNSTABLE
REACTION IN A 1/16-INCH WALL PLEXIGLAS TUBE



After Watson, et.al. (184)
Photos courtesy BuMines

Table 5 summarizes the results of the studies performed for nitroglycerin-ethylene glycol dinitrate systems.

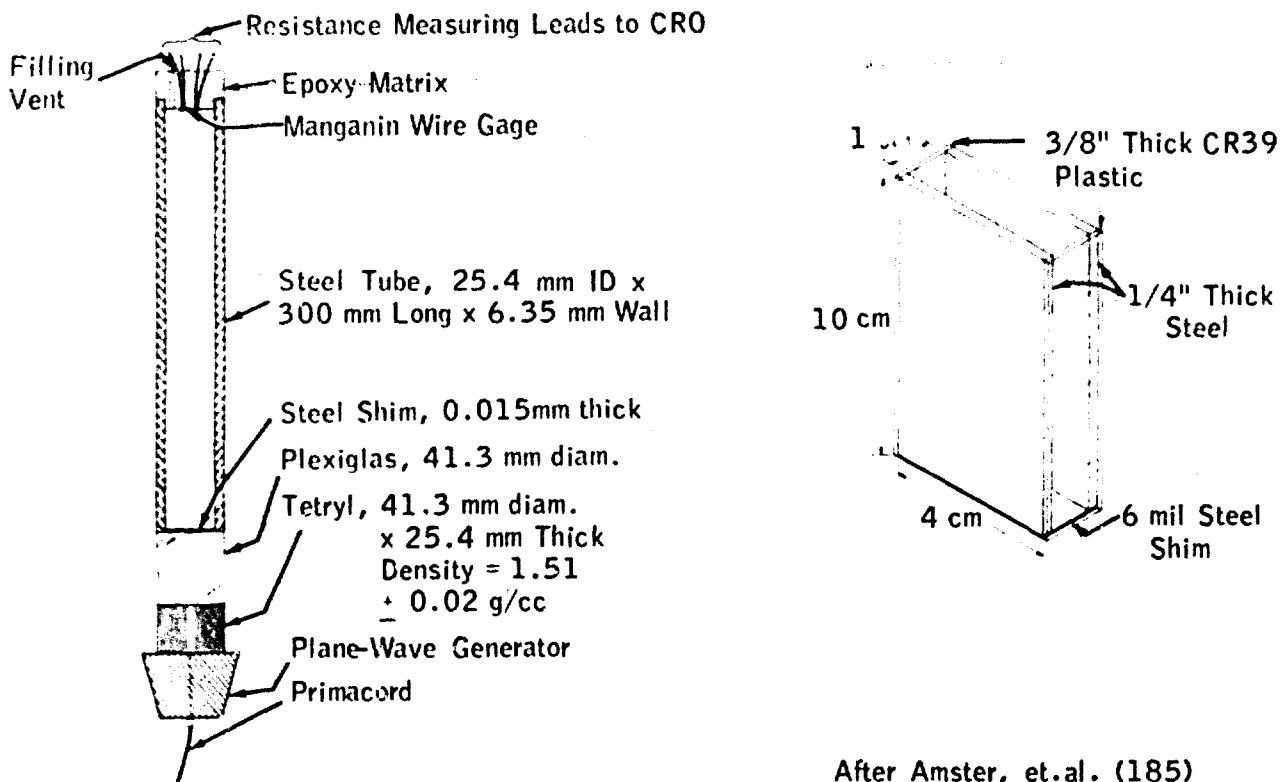
TABLE 5
STABILITY OF LOW VELOCITY DETONATION NG-EGDN

<u>Wall Thickness, In.</u>	<u>Result</u>	<u>Detonation Velocity, mm/μsec.</u>
<u>Lead Tubes; Co = 1.21 mm/μsec.</u>		
1/16	Unstable	----
1/8	Unstable	----
1/4	Unstable	----
<u>Plexiglas Tubes; Co = 1.84 mm/μsec.</u>		
1/16	Unstable	----
1/8	Stable	2.14
1/4	Stable	1.84
<u>Steel Tubes; Co = 5.20 mm/μsec.</u>		
1/16	Stable	1.96
1/8	Stable	1.88
1/4	Stable	2.11
<u>Aluminum Tubes; Co = 5.00 mm/μsec.</u>		
1/16	Unstable	----
1/4	Stable	2.04

These results indicate that the sonic velocity of the tube wall material and the wall thickness are the important factors in the propagation of stable low velocity detonations. Unstable reaction waves were observed in experiments in which the tube walls (lead) possessed a sonic velocity less than that of the liquid explosive, for all wall thicknesses. Materials possessing sonic velocities in excess of that for the liquid explosive produced stable low velocity detonations except when very thin tube walls were employed. Thus, it is suggested that perturbations of the liquid explosive resulting from precursor wall waves exert a significant influence on the reaction wave phenomena which occur.

In the SRI experimental study (185), both high and low velocity detonations were observed for a liquid explosive system consisting of 90% nitromethane/10% tetranitromethane. Two experimental test geometries have been employed in testing this explosive system, as indicated in Figure 15.

Figure 15
SRI APPARATUS



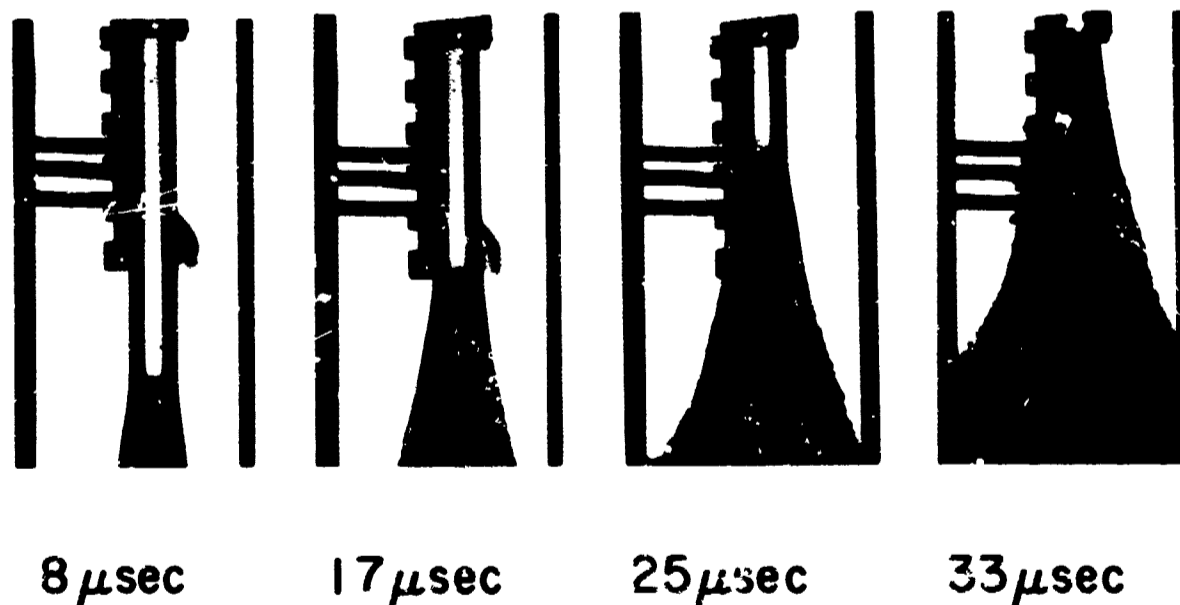
After Amster, et.al. (185)

The first test geometry consisted of a steel cylinder 300 mm long having an inside diameter of 25.4 mm and a wall thickness of 6.33 mm. The second fixture was a windowed sample container or rectangular cross-section, having two walls of 1/4 inch steel and two walls of 3/8 inch Plexiglas. The shock donor for both test geometries consisted of a Plexiglas attenuator coupled to a tetryl booster which was initiated by a plane wave generator. This initiating system is capable of producing input shock pressures up to approximately 60 kilobars.

In the experiments conducted with the cylindrical system, both high and low velocity detonations were observed. With input shock pressures of the order of 100 kilobars, high velocity detonations propagating at 6100 to 6400 m/sec. were initiated. Entering pressures below about 60 kilobars initiated stable low velocity detonations which propagated at approximately 1700 m/sec. Analogous experiments conducted with pure nitromethane reliably yielded high velocity detonations when initiating shocks were approximately 100 kilobars or above; but no low velocity phenomena were observed with pure nitromethane when input pressures were reduced, suggesting that nitromethane will not undergo low velocity detonation.

Experiments conducted with the windowed metal cell apparatus failed to produce low velocity detonation even though the input shocks were of a magnitude appropriate to the initiation of such phenomena, but high velocity detonations were readily initiated under the usual conditions. Figure 16 illustrates a high velocity detonation in this apparatus. In this figure, the advancing shock wave and material expansion can be clearly seen.

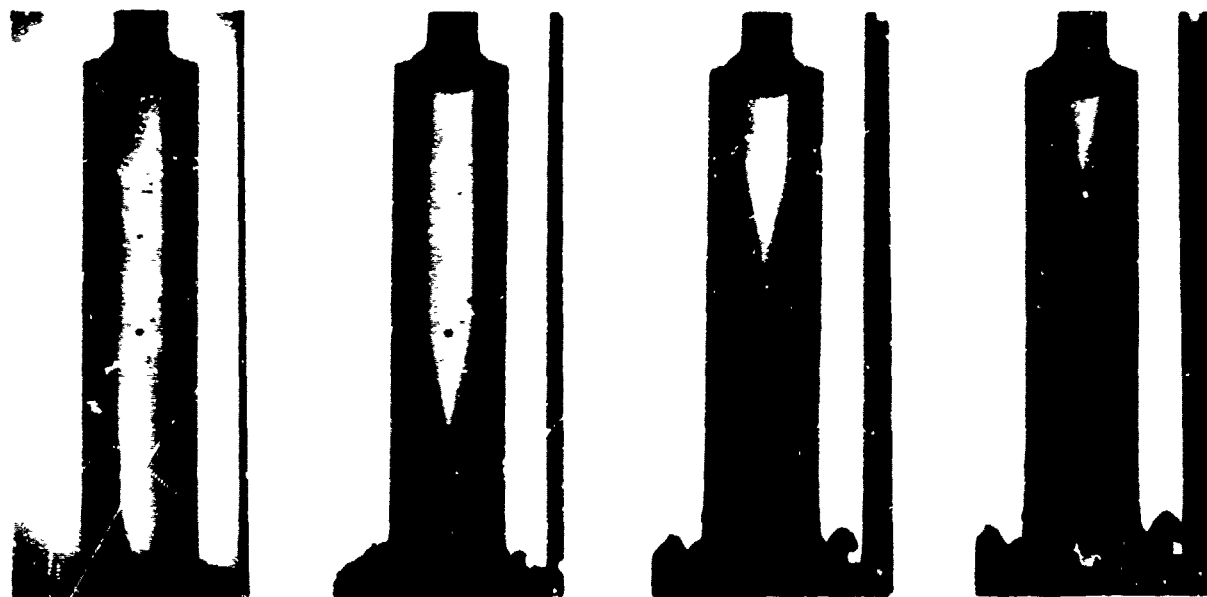
FIGURE 16
FRAMING CAMERA PHOTOGRAPHS OF HIGH
VELOCITY DETONATION IN 10% TNM - 90% NM



After Amster, et.al. (185)
Photos courtesy Stanford Research Institute

Figure 17 illustrates an attempt to initiate low velocity detonation. The interacting bow waves in the liquid explosive can be easily seen, although no chemical reaction was initiated.

FIGURE 17
FRAMING CAMERA PHOTOGRAPHS OF SHOCK
PROPAGATION IN 10% TNM - 90% NM
CONTAINED IN 1/4 INCH STEEL WALLS



0 μ sec

6 μ sec

11 μ sec

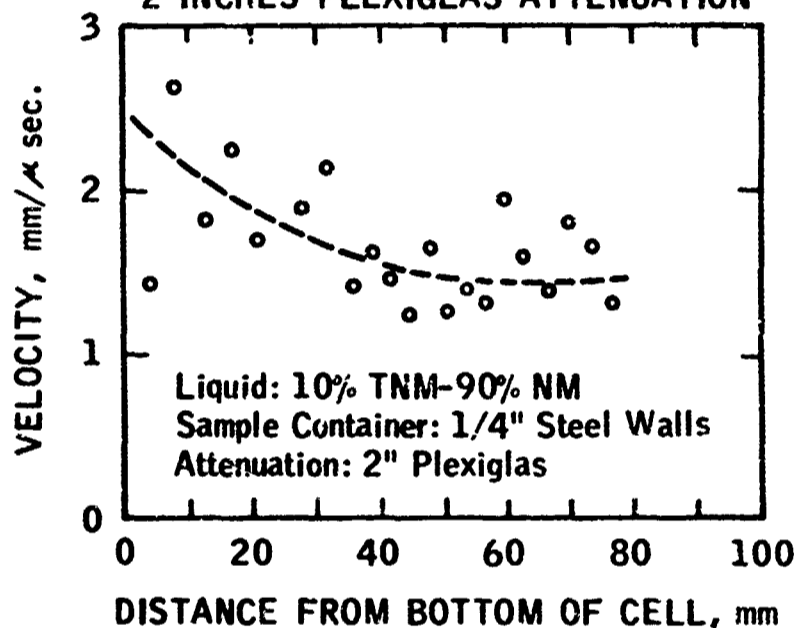
14 μ sec

After Amster, et.al. (185)
Photos courtesy Stanford Research Institute

Figure 18 provides a graphical representation of the velocity-distance behavior of the leading dark waves seen in Figure 17.

Figure 18

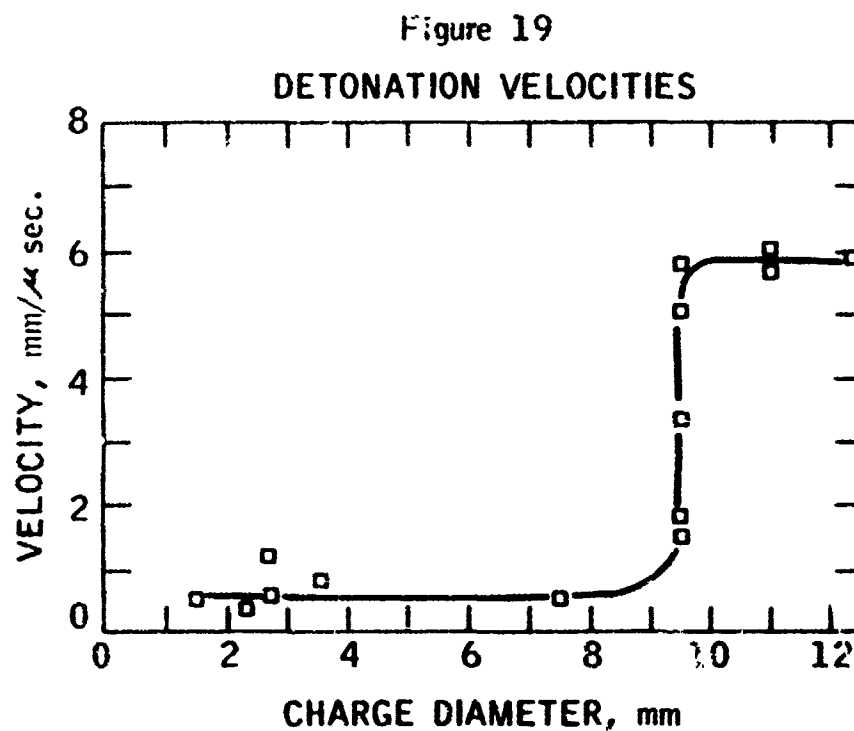
SHOCK VELOCITY IN 10% TNM-90% NM IN WINDOWED
SAMPLE CONTAINER: 1/4 INCH STEEL WALLS,
2 INCHES PLEXIGLAS ATTENUATION



After Amster, et.al. (185)

The propagation velocity of these waves is that of low velocity detonation in the cylindrical geometry. The fact that low velocity wave phenomena could not be initiated in the rectangular cross-section system even though the precursor bow waves are apparent suggests that some wave interaction in the cylindrical system is responsible for the initiation and propagation of low velocity detonation.

In a different investigation at Stanford Research Institute, Seely found that certain (classified) substituted butane explosives exhibited two distinct detonation velocities, depending reliably upon the diameter of the charges. At a recent symposium (186), Seely reported fourteen experimental determinations as follows:



After Seely, et.al. (186)

These compositions clearly detonate in the intermediate range in diameters under 9 mm. The confinement was heavy-walled lead, and initiation was by an RDX pellet.

It can be therefore seen that certain liquid explosives exhibit the phenomenon known as LVD while others, notably nitromethane, do not. The severity of initiation and the sonic velocity, geometry, and thickness of the confinement apparently exert a significant influence on the LVD phenomena, but the details of the interacting mechanisms have not been established. It is apparent that the chemical properties of the explosive also play a strong part in the LVD phenomenon.

Granular Explosives

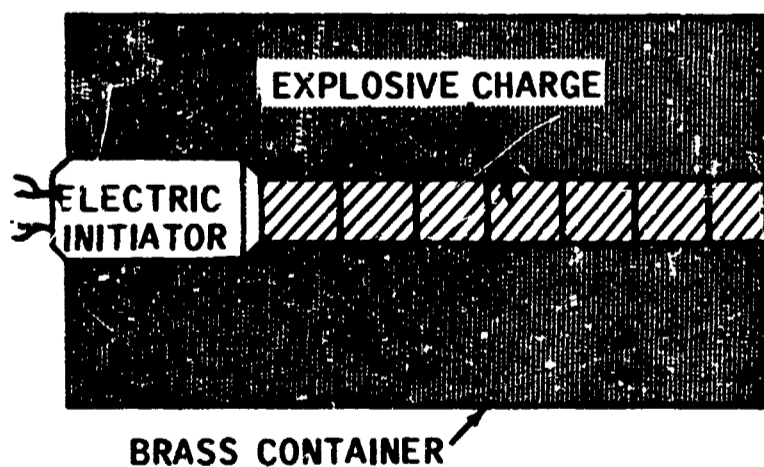
Experimental data concerning observation of multiple detonation velocities exists for granular explosive systems, although fewer studies have been made than for liquid systems. As with liquid explosives, granular materials, under appropriate conditions have been observed to detonate at two distinct velocities; and the main factor controlling the propagation regime again appears to be severity of initiation.

Jones and Mitchell (187) observed low velocity detonations with loosely packed granular TNT and tetryl. Flake TNT cartridged at a density of 1 gm/cc and initiated with a #6 detonator propagated at 1120 m/sec. The same charge when initiated with a 12.5 gram tetryl pellet produced a detonation velocity of 3660 m/sec. Tests performed with charges up to one meter in length showed no significant change in detonation velocity with charge length. In this study, it was noted that the low velocity detonation regime was characterized by incomplete decomposition of the explosive. The presence of undecomposed explosive in the residue was confirmed by chemical tests and a significant portion of the explosive was scattered without decomposition.

Stresau (188) has studied low velocity detonations in primary explosive systems. The experimental apparatus is shown in Figure 20.

Figure 20

APPARATUS FOR STUDY OF LVD IN PRIMARY EXPLOSIVES

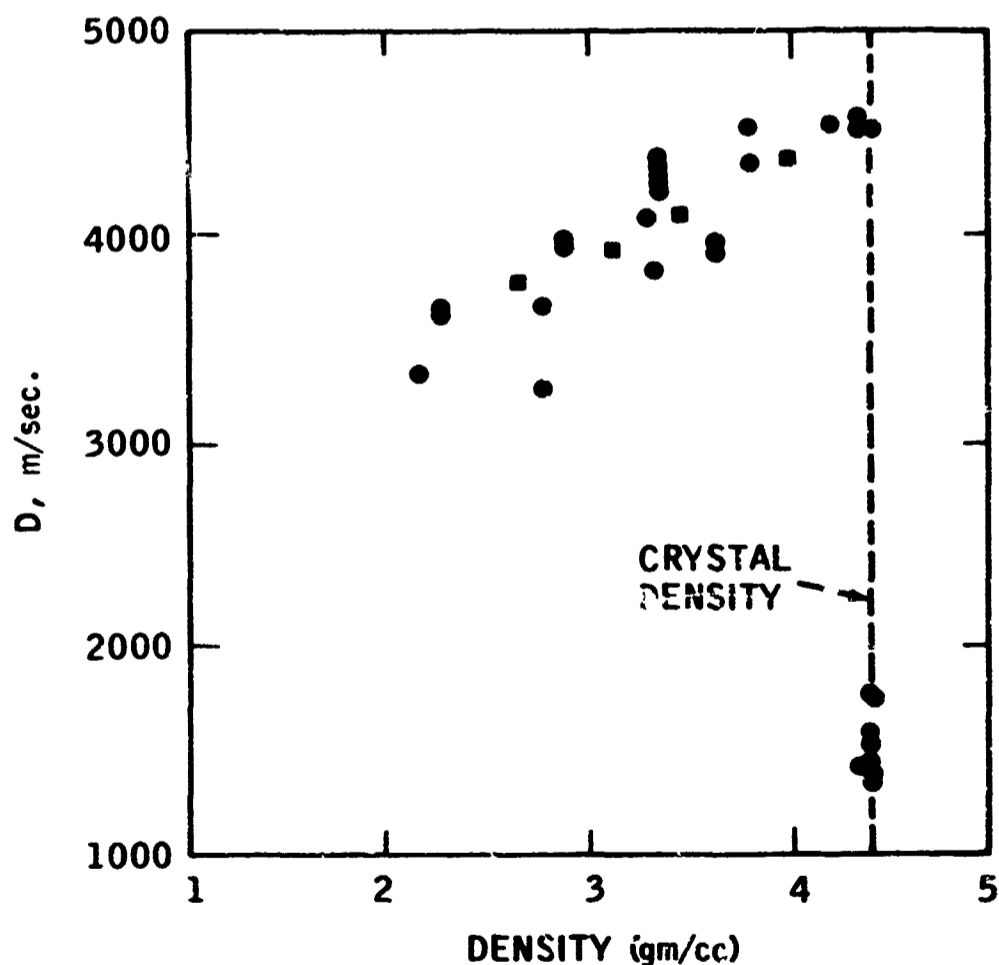


After Stresau (188)

The explosives were loaded into the tubes in increments of not more than one diameter in length to insure constant density; and electric initiators were used. In this effort, lead azide and mercury fulminate were observed to detonate at velocities of 1400 to 1700 m/sec. under heavy confinement. The normal detonation velocity of these materials is approximately 5000 m/sec., and this regime could also be obtained in the experimental apparatus under proper conditions of loading and initiation.

The results of this study differ from those of others in one important aspect. Low velocity detonations could only be observed when the charge was loaded at very high density. In fact, the charges had to be loaded at pressures such that the interstitial voids accounted for less than 6-7% of the volume to obtain low velocity detonation. Loading to lower densities resulted in high velocity detonation agreeing with normal thermohydrodynamic theory. Figure 21 presents the experimental data pertaining to mercury fulminate.

Figure 21
VELOCITY OF PROPAGATION VS.
LOADING DENSITY
0.150 DIA. COLUMNS



After Stresau (188)

These data indicate the proper theoretical variation of detonation velocity with loading density up to approximately crystal density. At crystal density, weak initiation leads to apparently stable low velocity detonation.

Low velocity detonation in granular charges of secondary explosives has been studied by Gurton (182). In this work, the effects of initial pressure and interstitial gas species were explored in granular charges of TNT, tetryl, and nitroguanidine. The charges were prepared by loosely packing the granular materials into perforated paper or celluloid cylinders 3/4 inch in diameter and 5 inches long. Packing densities were 0.9 gm/cc for tetryl, 0.8 gm/cm³ for TNT and 0.5 gm/cm³ for nitroguanidine. Before firing, the charges were evacuated and repressured with the gas under study. A summary of the experimental results obtained using granular tetryl is presented in Table 6. Similar results for TNT and nitroguanidine are presented in Tables 7 and 8.

TABLE 6

EFFECT OF PRESSURE ON THE VELOCITY OF DETONATION OF GRAINED TETRYL
DENSITY 0.9 g/cc SET OFF BY A NO. 6 A.S.A.T. COPPER DETONATOR

<u>Diameter</u> <u>Inches</u>	<u>Pressure</u> <u>Atmospheres</u>	<u>Gas Filling</u> <u>Voids</u>	<u>Velocity of Detonation</u> <u>m./sec.</u>
7/16	0.03	Air	1,460*
7/16	1.0	Air	1,480, 1,300, 1,330 1,430*, 1,540*
7/16	14.3	Methane	910
7/16	27.7	Methane	Failed
3/4	1.0	Air	1,700
3/4	14.3	Methane	1,890
3/4	21	Methane	1,450
3/4	27.7	Methane	1,330
3/4	47.7	Methane	Failed
15/16	1.0	Air	2,860
15/16	14.3	Methane	2,330
15/16	17.6	Methane	2,085
15/16	21.0	Methane	1,695
15/16	41.0	Methane	Failed

* Determined by high speed camera method.

TABLE 7

VELOCITY OF DETONATION OF T.N.T. FLAKE DENSITY 0.8 g/cc
FIRED BY A NO. 6 A.S.A.T. COPPER CASED DETONATOR

<u>Diameter Inches</u>	<u>Pressure Atmospheres</u>	<u>Gas Present</u>	<u>Velocity of Detonation m./sec.</u>	
3/4	0.03	Air	2020*	2020*
	0.12	Air	1754	1818
	1.0	Air	1750	2040
	21	Methane	2080	
	24.3	Methane	No reading obtained	
	27.7	Methane	Some T.N.T. left	
	41	Methane	Failed completely	
5/16	1.0	Air	2,380	
	9.3	Methane	2,330	
	11.0	Methane	1,020	
	14.3	Methane	2,000	
	26	Methane	1,820	
	41	Methane	Failed completely	

* Determined by high speed camera method.

TABLE 8

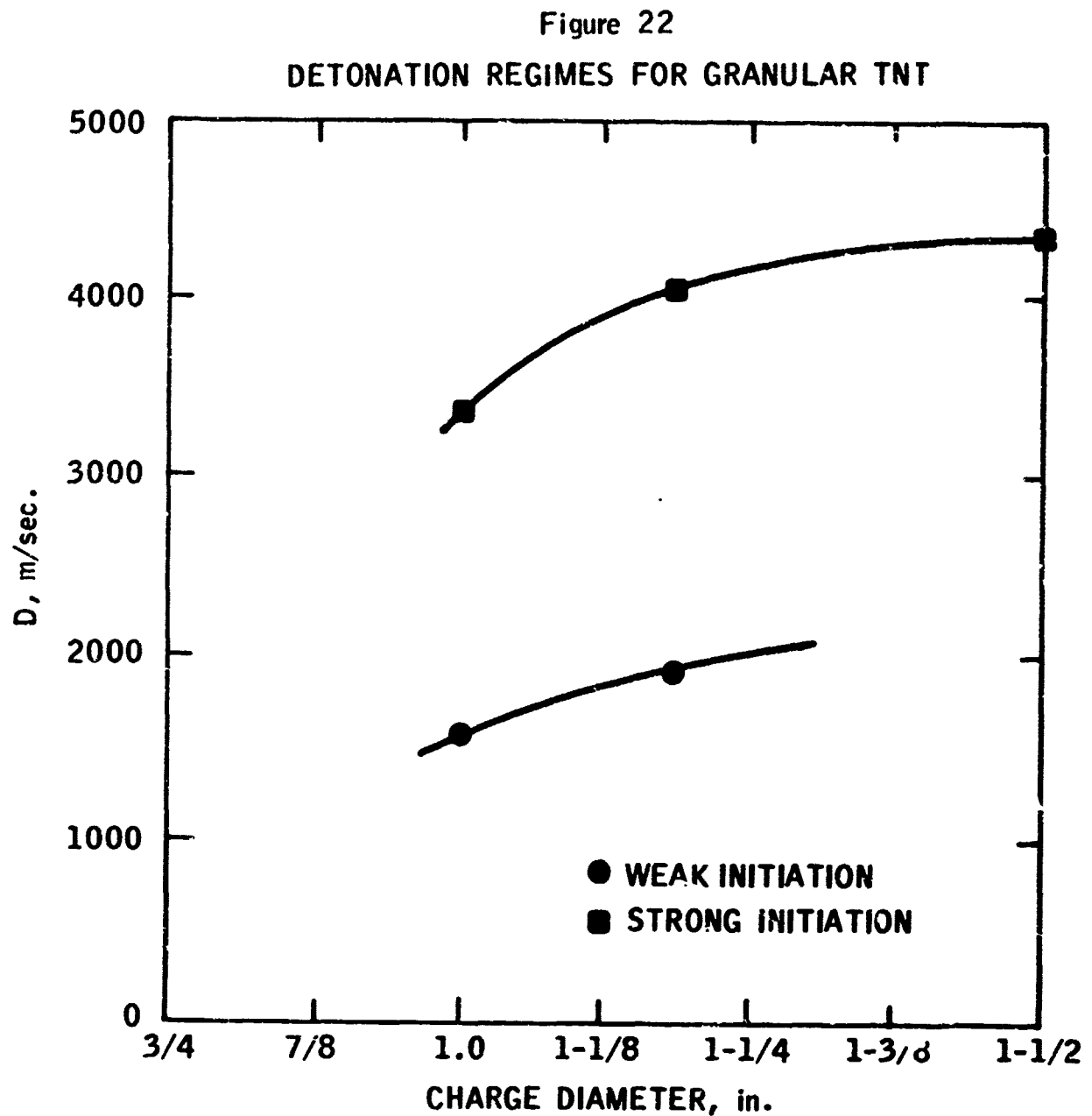
DETONATION VELOCITY OF NITROGUANIDINE DENSITY 0.5 g/cc
SET OFF BY A NO. 6 A.S.A.T. COPPER CASED DETONATOR

<u>Diameter of Cartridge</u> <u>Inches</u>	<u>Pressure</u> <u>Atmospheres</u>	<u>Gas</u> <u>Injected</u>	<u>Velocity of Detonation</u> <u>m./sec.</u>
7/16	0.03	Air	2650*
	1.0	Air	2750 2680* 2700*
	6.0	Methane	2500
	7.0	Methane	1490
	11.0	Methane	2170
	17.7	Methane	Failed
3/4	1.0	Air	3130
	6.7	Methane	3220
	11.0	Methane	3030
	14.3	Methane	Failed

* Determined by high speed camera methods.

In all cases, the charges were initiated with #6 detonators. The experimental data indicate that the ease of propagation of low velocity detonation is reduced as the interstitial gas pressure is increased. A threshold pressure exists above which the detonation does not propagate. High velocity detonations were not affected by interstitial gas pressure and propagated normally once initiated. Thus, it appears from this study that the presence of interstitial gas in granular charges of secondary explosives plays a significant role in the initiation and propagation of low velocity detonation.

In a study of detonation sensitivity, Jones and Cumming (189) observed two detonation regimes for granular charges of TNT. The experimental results are presented in Figure 22.



After Jones and Cumming (189)

This study noted a significant effect of charge diameter. Charges 1 inch in diameter, which were initiated at any velocity between 1600 and 3200 m/sec., accelerated or decelerated until the detonation rate reached approximately 1700 m/sec., where it stabilized. If such charges were initiated at less than 1600 m/sec., the detonation died out. Initiation at above 3200 m/sec. led to a stabilized detonation rate of 3400 m/sec. Similarly, two regimes were noted for charges 1-3/16 inch in diameter. The initiation limits were slightly modified for such charges. Only one stable velocity was noted for 1-1/2 inch diameter charges: 4400 m/sec.

Low velocity detonation propagation in certain solid secondary explosives has been recently studied in Russian laboratories (190). Explosive specimens 1 mm thick and 50 mm in diameter were compressed between metal and Plexiglas cylinders and initiated with a hot wire initiator. The events were registered through the Plexiglas cylinder using high speed photographic techniques. The detonation was found to start as a slow burning which propagated rapidly in the radial direction. The expanding reaction products caused an increase in pressure creating a compression wave which propagated in front of the flame at sonic velocity. A weak detonation wave was found to form behind the compression wave, and this initiated reaction at sites of nonuniformity. The detonation process was found to stabilize upon reaching certain velocities, which were 700-800 m/sec. for PETN, 700 m/sec. for hexogen, 550 m/sec. for DINA, and 300 m/sec. for tetryl. The detonation pressures for PETN and hexogen were calculated to be 3.8 and 3.0 kilobars, respectively. The reaction zone, under the conditions studied, was markedly longer than usual for these materials.

The propagation of low velocity detonations in powdered trotyl, tetryl, and hexogen has been studied photographically (191), using both weak and strong initiators. This work showed that, with weak initiation, a range of charge diameters existed within which the low velocity detonation was stable for a time, with detonation velocity increasing from 1200 m/sec. to 1800-2200 m/sec. as the charge diameter was increased. In tetryl and hexogen, the low velocity detonation was found to transit to normal detonation after a run of from 2-4 charge diameters. The transition was more gradual in trotyl charges. The use of strong detonators led to normal high velocity detonations.

Hershkowitz (192,193,194) has studied the propagation of reaction waves in granular mixtures of potassium perchlorate and aluminum. In this study, stable reaction waves were observed to propagate at either a low velocity of 300 m/sec. or a high velocity of about 900 m/sec. The lower velocity regime was observed only in a few cases. The theoretical detonation velocity for such mixtures is approximately 5000 m/sec., but such velocities have not been obtained in the experimental apparatus. Calculations concerning the penetration depth from the combustion front into the mixture for diffusion of mass and energy suggest that a deflagration would propagate at less than 0.3 m/sec., and the calculated sonic velocity for the mixture was less than 300 m/sec. Considering these factors, it was concluded that the experiments illustrated detonative phenomena rather than deflagrative phenomena. Other (classified) work with metal/metal perchlorates has given detonation rates of about 3000 m/sec.

Hershkowitz's charges were prepared by pouring five-gram increments of 60/40 mixtures of potassium perchlorate/atomized aluminum into a 3/8 inch diameter axial hole drilled into a 3-inch diameter Lucite cylinder 20 inches in length. Experiments were also conducted in steel and aluminum confinements. Initiation was accomplished either by tetryl pellet or by zirconium squib. The events following initiation were observed with both streak and framing cameras.

Cook (U.S. Patent 2,199,218) and Davis (U.S. Patent 2,168,562) have described systems which detonate at velocities as low as 1100 m/sec. These systems consist of granular ammonium nitrate, thinly coated with explosives such as nitroglycerine, TNT, tetryl, or HMX. The purpose of these systems was sensitization of ammonium nitrate to intentional initiation. It is not clear why such low detonation velocities were obtained.

Gates (U.S. 3,299,811) has patented a series of solid mixtures which detonate at the relatively low rates of 1250 m/sec. to 2436 m/sec., with the production of very little gas. They are mixtures of high explosives, aluminum and metal perchlorates. Typical data from Gates' patent are given in Table 9.

TABLE 9
PROPERTIES OF EXPLOSIVE COMPOSITIONS

<u>Material</u>	<u>Composition</u>	<u>Detonation Velocity, m/sec</u>
Mild Detonating Fuse	100% PETN	7300
Pyrocore	20% PETN 40% Al 40% BaO ₂	4200
Slowpoke 32	20% PETN 32% Al 48% KClO ₄	2000
Slowpoke 44	20% PETN 44% Al 36% KClO ₄	1430
Slowpoke 56	20% PETN 36% Al 24% KClO ₄	1350
Calpoke	20% PETN 55% Al 25% Ca(ClO ₄) ₂	1250

The observed LVD phenomena in granular systems thus appear to be dependent both upon charge diameter and upon interstitial gas composition. The strong diameter dependence suggests that lateral losses might play a significant role in such behavior. The interstitial gas properties could have an important effect on the initiation phase of the overall reaction process. It must be realized that the granular explosives differ from the liquid explosive systems in that they are quite inhomogeneous, and interactions of shock waves and density discontinuities can create local initiation sites.

The observation of apparent LVD phenomena in primary explosive systems cannot be explained with any of the existing ideas concerning LVD. The observations reported here are apparently the only ones which have been made. More experimental data are necessary before conclusions can be drawn.

The explosive systems containing aluminum are subject to rate limitations as a result of physical kinetic processes, such as vaporization. The observed LVD phenomena are most likely the result of complex interactions between such physical kinetic processes, diameter effects, and effects of inhomogeneity. The initiation characteristics of such systems are extremely complex and difficult to unravel.

Convective deflagration

Four phenomena have been observed which appear to be ultra-fast, probably convective, deflagration; two of these are new.

The rapid combustion of gunpowder in an artillery or shot-gun shell is too well-known to require description except to point out that the gunpowder is in the form of loosely-packed granules with voids through which a flame can flash when driven by the high pressure of the heavily confined combustion. The entire powder charge is ignited effectively at once by the pressure-driven flame, and the reaction zone fills the entire shell as each individual granule proceeds to burn. The bulk burning rate is high due to the high pressure, but no higher than predicted by the normal pressure exponent law. The rate of advance of the combustion front through the granular charge, however, is very much higher. The essential features of the phenomenon are (1) high porosity and (2) heavy confinement.

Very rapid burning fuses have been made (chiefly by the British) by filling heavy tubes with black powder perforated with longitudinal channels. The heavy confinement forced the flame front down the channels at high rates of speed. Black powder is especially amenable to this application because of its high ignitability and its nondetonability (double-base is detonable). The fuses never proved very useful because their rates depended so heavily on the generated pressure, which in turn depended upon the confinement; so that reproducibility of action times, with reasonable constructions, was prohibitively poor.

The McCormick-Selph Company reports that they have developed a family of (proprietary) explosives which propagate with true, reproducible, intermediate rates. Their work has not been published, but the manager of McCormick-Selph's Explosives Division gave the following information (195):

These explosives are based upon a family of pure compounds which are virtually gasless monopropellants and which yield approximately 450 cal./gm. The pure compounds burn at approximately 4.5 m/sec. in long, thin columns or in 0.5 inch diameter pressed pellets. The addition of an inorganic oxidizer (up to 85 wt.%) increases the heat yield (up to 1500 cal./gm.) and the gas yield (up to somewhat more than smokeless powder). The propagation rates of oxidized compositions range up to 280 m/sec. Stoichiometrically oxidized compositions give no smoke and no residue. The parent compounds are stable to 370°C for at least 24 hours. The parent compounds currently cost approximately \$1800-2000/lb., synthesized on a laboratory scale. It is estimated that they would cost \$200-500/lb. in tonnage lots.

The pressures generated appear to be in the intermediate range. Fuse cords made from these materials will not initiate PETN, which implies a shock pressure of ≤ 2 kbar. An accidental explosion of 1 pound of oxidized mixture on a 1/4 inch steel plate produced a dent approximately 3 inches deep. Experienced observers felt that black powder (nondetonating) would have produced no dent, and that an ordinary high explosive would have punched a clean hole. Full-scale card-gap tests have not been run; however, initiations of columns and/or pellets by squibs, blasting caps, Primacord, and/or a 1/2 inch thick stack of RDX sheet explosive have always given deflagrations as described above, and never detonations.

All the work on this system has been done in one laboratory, and detailed studies of the propagation mechanism have not been made. However, it is known that the compositions are porous solids, with high melting points, and they appear to be nondetonable; so the essential conditions for convective deflagration appear to be present.

It has also been reported (206) that granules of Pyrofuse (palladium-coated aluminum) will "burn" at bulk velocities up to 250 cm/sec. The reaction is one of alloying, not of combustion; and the reaction product is solid palladium-aluminum alloy. The heat of reaction is sufficient to boil the alloy, so a gas is present during reaction and could well be the heat transfer medium. Like the preceding example, this bimetallic composition is porous, nondetonable, thermally stable, gasless (after cooling), and has a rather high heat yield per gram.

2.3.2. Mechanisms Which Have Been Advanced

No comprehensive theory applicable to intermediate velocity reaction wave phenomena has yet been developed, but there have been many attempts to explain individual phenomena or classes of phenomena. In Section 2.3.1., the observed phenomena were grouped into four categories:

- Ideal detonation in low-density media
- Nonideal detonation
- LVT in liquids and solids
- Ultra-fast (probably convective) deflagration

Of these groups, the first is well understood via thermohydrodynamic theory and needs no discussion beyond that given above; the other three are only incompletely understood. The approaches to the detonative phenomena can be broadly categorized into two classes: those treatments which relax the basic assumptions in the Zeldovich/von Neumann/Döring theory (which are that the flow is one-dimensional and that all the chemical reaction energy contributes to sustaining the detonation wave), and those treatments which consider that the intermediate phenomena are due to multi-dimensional effects and cannot be explained within the framework of the one-dimensional theory at all. Ultra-fast deflagration is only qualitatively understood.

Nonideal Detonation

Detonations become nonideal when appreciable amounts of reaction energy are lost because significant lateral expansion takes place between the initiating shock and the Chapman-Jouguet plane. Theories pertaining to lateral expansion have been developed by Jones (51), Eyring and co-workers (52), and Cook (53).

In the theory developed by Jones (51), the reaction is visualized as starting at a plane shock front and ending at another plane a distance "a" behind. Thus, the distance "a" is the length of the reaction zone. Since the coordinate system is moving with the shock front, the products of detonation appear like material issuing from a nozzle, the mouth of the nozzle being at the wave front. For this reason, the Jones Theory has been called "nozzle theory." In the Jones development, it was considered that lateral expansion had a negligibly small effect on the hydrodynamic equations expressing conservation of energy and momentum. The perturbed equation of continuity was written as

$$r^2 \rho_{(D-u)} = \rho_o^D \quad (10)$$

where r is the relative expansion of the central stream tube along the reaction zone. Using this expression, a constant co-volume equation of state, the unperturbed conservation equations for energy and momentum, and an expression for the ideal detonation velocity in a gaseous system, Jones obtained the expression

$$(D_i/D)^2 = 1 + 9/4 (r_1^4 - 1) \quad (11)$$

relating the detonation velocity to the ideal velocity and to the relative expansion of the central stream tube, r_1 , at the end of the reaction zone.

In order to obtain the relative expansion of the central stream tube, Jones assumed that the gases flowing from unconfined charges follow the flow lines given by Meyer's solution for flow around a corner. He further assumed that the gases from the central portion of the explosive expand at constant pressure in any given cross section. By joining the two solutions along an appropriate streamline, the final equations determining the expansion as a function of the reaction zone length become

$$r_1 = \frac{37}{20} (1 - a/R \cot \theta) \quad (12)$$

$$\frac{a}{R} = \frac{34}{37} \frac{\sin \theta}{1 + \cos \theta} \sqrt{2\theta} \quad (13)$$

Thus, actual detonation velocity has been related to the ideal detonation velocity through the reaction zone length and the charge radius. The above equations indicate that the effect of the finite reaction zone length begins to become significant when the reaction zone length approaches the radius of the charge.

The curved front theory of Eyring and co-workers (52) also assumes that only the equation of continuity is perturbed by lateral expansion. It was reasoned that a rarefaction wave would be sent into the reaction zone from the edge of the charge; and, since the local velocity of sound in the reaction zone is greater than the detonation velocity, the detonation wave would be overtaken by the rarefaction wave and slowed down near the edges of the charge. Thus, the detonation wave front would appear curved and the detonation velocity would be less than ideal because of the curvature. It was considered that a steady state would be achieved when the angle of the inner section of the wave front with the edge of the charge is sufficiently small so that the rarefaction wave is no longer reflected. The equation of continuity was written

$$(D-u) \frac{dV}{dr} = \frac{2uV}{r} - V \frac{du}{dr} \quad (14)$$

$$D = D_0 \cos \phi \quad (15)$$

which follows from the assumption that, to a first approximation, any small portion of the wave front may be considered spherical, with a local radius of curvature, r_0 . D is the velocity normal to the wave, and is given by Equation 15, being the angle between the axis of the charge and the normal to the wave at any point. In this fashion, the normal detonation velocity can be calculated from the perturbed hydrodynamic equations. Employing the expression for calculating the ideal detonation wave velocity, it is possible to obtain an expression for the actual detonation velocity in terms of the reaction zone length and the charge radius. The final result is

$$\frac{D}{D_i} = 1 - 0.5 \left(\frac{a}{R} \right) \quad (16)$$

The curved front theory permits theoretical calculation of the actual wave shape. Such calculations indicate wave fronts which decrease sharply in radius of curvature from the central axis to the periphery of the charge, which is in qualitative agreement with experimental observation.

Wood and Kirkwood (196) analyzed the relation between the velocity and radius of curvature of the wave front and the charge radius. Their equation for the detonation velocity is sensitive to the Chapman-Jouguet pressure and the von Neumann spike compression, and is dependent upon the equation of state. Green and James (197) have recently used this result to experimentally determine average reaction zone thicknesses for several explosives.

Cook (53) developed a theory of nonideal detonation, known as the "geometrical model," in which the detonation velocity is determined by the quantity of chemical reaction energy released ahead of a critical region within the reaction zone. As the external confinement becomes less severe, the critical region moves closer to the shock front. If the reaction zone length is smaller than the distance from the initiating shock to the Chapman-Jouguet point, the critical region will obviously lie within the reaction zone as defined in ideal detonation theory; and the case degenerates to ideality. If, however, the critical region lies beyond the Chapman Jouguet point, only a portion of the chemical reaction energy is applied to sustaining the detonation; and the situation becomes nonideal. In essence, then, this becomes a treatment of effective reaction zone length.

Quantitative calculations using the geometrical model can be made only when an exact reaction rate expression has been obtained for the decomposition. Employing Eyring's surface burning model for granular materials, Cook obtains

$$D/D_i = \left\{ 1 - [1 - 4h(L,d)/3D\tau]^3 \right\}^{1/2} \quad (17)$$

in which h represents the length of the critical region. h is obtained from detonation head theory by the relationship:

$$h(L,d) = \begin{cases} (L/3.5d)d_{eff}; & L < L_m \\ d_{eff} & ; L \geq L_m \end{cases} \quad (18)$$

The experimental data for any one explosive are not sufficiently precise to decide which of the above discussed theories most closely represents physical reality; but, on balance, the nozzle theory and the geometrical model appear to give the best agreement with experimental data. Figure 10 (page 49) presents data measured by Anderson (170) for detonation in ammonium perchlorate, and the results of application of the three theories are shown. In this particular case, it appears that the data are best represented by the curve computed using the geometrical model determined by Cook.

The simplifying assumptions invoked to produce tractable equations pertaining to "nonideal" detonation strongly limit the accuracy with which the phenomena can be predicted. Such theories are not particularly helpful in understanding the actual physical/chemical interactions which determine the characteristics of the observed reaction waves.

LVD in Liquids and Solids

LVD, or Low Velocity Detonation, is at present a poorly defined phenomenon. It occurs, under special conditions of confinement and/or initiation, in systems which can also undergo a normal, high velocity detonation; and the special conditions are critical. The LVD mode also (usually) easily transits to a high velocity detonation. The attempts to explain LVD all consider that LVD is dependent upon multi-dimensional wave interaction phenomena and so cannot be interpreted within the framework of the one-dimensional theory.

Fyring and co-workers (52) began the development of the theory of detonation failure by assuming that the reaction zone length appearing in Equation 16 (above) is not a constant, but is in fact dependent upon the detonation velocity. This assumption, of course, will be justifiable only when the reaction zone length and the detonation velocity are obtained by noting that the reaction zone length is proportional to the reaction time, and the reaction time is the reciprocal of a specific rate constant whose temperature dependence can be determined as

$$a = \frac{\rho_0}{\rho_1} D \tau \quad (19)$$

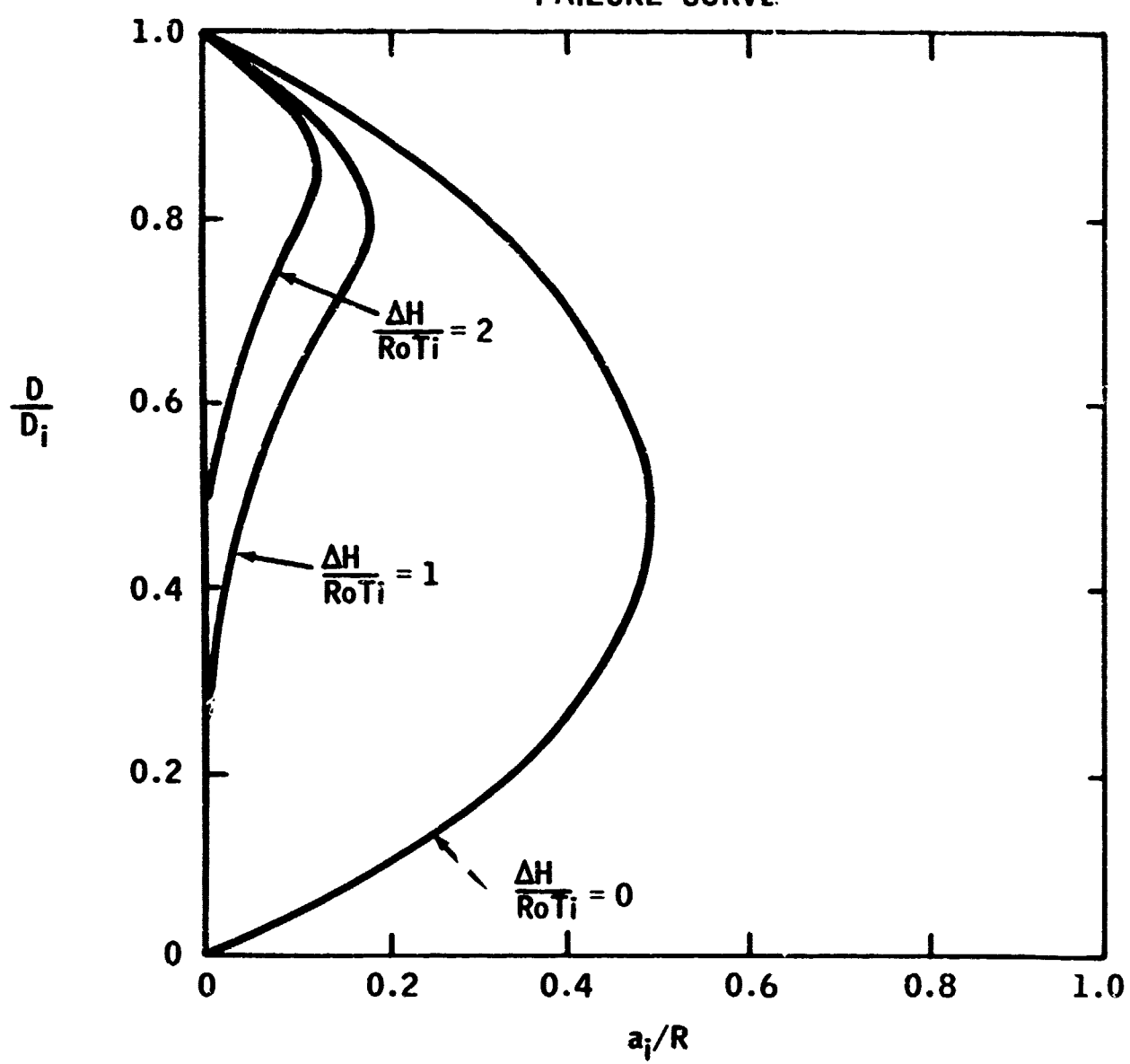
$$\frac{1}{\tau} = \frac{kT}{h} e^{\frac{\Delta S}{R_0}} e^{-\frac{\Delta H}{R_0 T}} \quad (20)$$

These expressions, when combined with the fact that temperature is dependent upon the square of the detonation velocity, yield

$$\frac{a}{a_1} = \frac{D_1}{D} e^{\frac{\Delta H}{R_0 T_1}} \left[\frac{D_1^2}{D^2} - 1 \right] \quad (21)$$

Using Equations 21 and 16, a plot of relative detonation velocity versus reaction zone length can be made for any desired value of the heat of activation. Figure 23 indicates such a plot for various values of the heat of activation.

Figure 23
FAILURE CURVE



After Eyring, et.al. (52)

Several conclusions can be drawn from Figure 23.

- A minimum radius exists, below which no stable detonation will propagate. Regardless of the severity of initiation, the detonation will fail when the radius of the charge is less than this minimum value.
- The critical radius and the corresponding critical velocity depend only upon the heat of activation.
- There can be two steady velocities for any charge radius greater than the critical radius. The higher of the two velocities corresponds to normal high order detonation, and imposed initial velocities higher or lower than normal will build up to or down to the steady state value. The lower velocity regime appears to be metastable such that imposed initial velocities of smaller magnitudes than those given by the lower branch lead to failure while higher velocities than those given by the lower branch build to high order detonation.

The theory predicts that low velocity detonation should propagate at lower velocities as charge diameter increases, an effect which has not been observed experimentally. It should also be noted that experimental observation of low velocity detonation has shown that LVD can occur in charges of less than the critical diameter, a consequence not allowed by the failure theory. In addition, the failure theory is based on the detonation reaction reaching energetic completion, while experimental observations of low velocity detonation suggest that this is not the case. Thus, while the failure theory does correctly indicate two velocity regimes; few, if any, of the other theoretical predictions are borne out by experimental observation. It is nevertheless tempting to assign the observed low velocity detonations to the lower branch of the failure curve indicated in Figure 22.

A similar theory has been developed by Evans (198). In this analysis, the expression for the detonation velocity derived by Wood and Kirkwood utilizing the radius of the curvature of the detonation wave is used with a single first order rate process description and the Hugoniot equation of state to calculate steady detonation wave solutions. It is found that, for a diameter less than a certain minimum, no solution is obtained. For larger diameters, the solution is dual valued, analogous to the result of Eyring. One major difference between this work and Eyring's lies in the fact that, in this work, the chemical reaction rate is determined from the shock temperature as opposed to the Chapman-Jouguet temperature.

Check calculations have been made to compare with available experimental data for liquid TNT and ammonium nitrate. These materials were selected because they have similar detonation velocities but quite dissimilar failure diameters. Calculations of failure diameter, detonation wave velocity at failure, induction times and shock sensitivities yielded results which compared relatively well with experimental values. Evans points out that Price

and Jaffey (199) reported an observation of detonation velocity for cast TNT of 5.23 mm/ μ sec. under conditions for which the propagation velocity should have been 6.8 mm/ μ sec.; it is possible that this experiment represents an example of a steady state wave on the low velocity branch of the curve.

The theory should not be expected to apply to heterogeneous systems because it implicitly assumes thermal decomposition in a homogeneous system. Any analysis which incorporates a reaction rate model having exponential temperature dependence will, in general, yield a dual-valued solution for detonation velocity in charges beyond the minimum diameter for wave propagation. In addition, the bulk of the experimental evidence of LVD indicates a much lower velocity for the low velocity regime than that corresponding to the lower state in the Evans theory. It is probable that the "lower" state in this theory is metastable and not observable experimentally.

The theory developed by Schall (200) differs from those of other investigators in that he considers that the total energy of the explosive is not released in, or at least does not contribute to the preservation of, the detonation wave. Only that amount of energy released between the shock front and the Chapman-Jouguet plane contributes to the propagation of the wave. Thus, if the entire reaction zone lies between the shock front and the Chapman-Jouguet plane, the total released energy contributes to the propagation of the wave and normal high velocity detonation occurs in agreement with the hydrodynamic theory; but if the reaction zone extends beyond the Chapman-Jouguet plane, part of the released energy is unavailable to drive the detonation wave. Schall considers that the latter is the case when an explosive is "under-initiated."

In the development of his theory, Schall considers that the distance from the shock front to the Chapman-Jouguet plane, a_1 , does not change with detonation velocity, and that the length of the reaction zone, a , is inversely proportional to the detonation velocity. Assuming that the chemical reaction energy is released uniformly throughout the reaction zone, that fraction of the energy released between the shock front and the Chapman-Jouguet surface becomes

$$Q = \frac{a_1}{a} Q_1 \quad (22)$$

where Q_1 represents the chemical reaction energy in an ideal wave. Using Equation 22 and the usual approximation that the liberated energy is proportional to the square of detonation velocity yields:

$$\frac{a_1}{a} = \frac{Q}{Q_1} = \left(\frac{D}{D_1} \right)^2 \quad (23)$$

The condition for wave stability is obtained by assuming that the chemical energy release resulting from an increase in the reaction rate due to an increase in the detonation wave velocity is less than the additional energy required to support a wave having the increased velocity. Equation 23 enables determination of the additional energy required to support the increased wave velocity:

$$\frac{dQ_w}{dD} = \frac{2Q}{D} \quad (24)$$

Using the assumption that the distance from the shock front to the Chapman-Jouguet plane is not a function of the detonation velocity, the increase in reaction energy release rate becomes

$$\frac{dQ_r}{dD} = \frac{dQ_r}{d(a_1/a)} \frac{a_1}{aD} \quad (25)$$

and the stability condition is obtained using Equations 24 and 25:

$$\frac{1}{2} \frac{a_1}{a} \frac{dQ}{d(a_1/a)} < Q \quad (26)$$

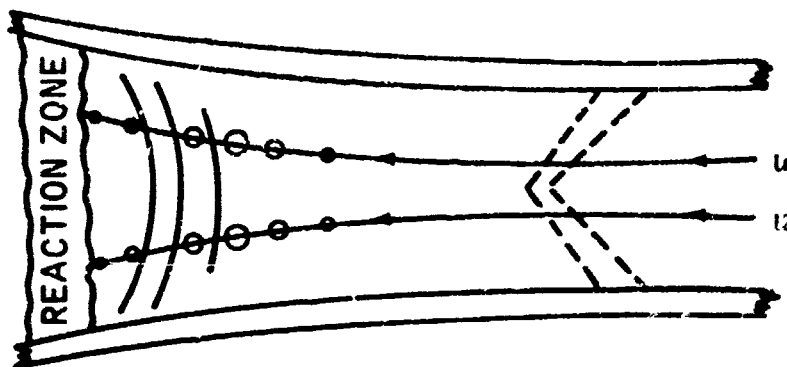
Schall considers a homogeneous, first order reaction scheme and has plotted the right-hand and left-hand sides of the inequality represented by Equation 26 versus the fraction completion of the reaction occurring between the shock front and the Chapman-Jouguet plane. This result indicates that a narrow, stable region exists for all values of the rate equation parameters when the reaction is essentially completed within the area between the shock front and the Chapman-Jouguet plane. A second, broader, stable region is indicated for certain values of the reaction rate expression parameters under conditions such that only a small fraction of the total reaction energy is released ahead of the Chapman-Jouguet plane. This result corresponds with the experimental observation that a range of low detonation velocities occurs rather than a single, well determined value. Schall also suggests that this type of analysis can be applied to explosive materials consisting of two or more components which possess very different reaction rates, and that stable detonation waves can result within whose Chapman-Jouguet plane only partial reaction takes place. An example of such a material would be an explosive containing aluminum.

Schall's assumption that the reaction zone length is inversely proportional to the detonation wave velocity is not in general true. The lengthening of the reaction zone for decreased wave velocity is associated with the ignition conditions at the wave front and is thus dependent upon the ignition mechanism. The detonation velocity corresponding to chemical

reactions completed ahead of the Chapman-Jouguet plane is influenced very little by the ignition mechanism, while the velocity range of the detonation waves corresponding to incomplete reaction is strongly dependent upon the ignition condition at the wave front. The Schall theory is very appealing in that it agrees qualitatively with experimental observations and none of its ideas are disproved by experimental observation. Insufficient experimental data are available, however, to test the theory quantitatively at this time.

Watson and co-workers at the Bureau of Mines (184) have taken a mechanistic approach in order to formulate a tentative hypothesis concerning their observation of low velocity detonation waves in liquid explosives. The model which they have proposed is illustrated in Figure 24.

Figure 24
BuMINES MODEL FOR LVD IN LIQUIDS



After Watson, et.al. (184)

Considering a coordinate scheme which moves with the reaction front, the sequence of events is described as a four-step process. In the first step, the undisturbed liquid explosive is compressed by bow waves in the liquid which are derived from a precursor wave in the container wall. In step 2, compressed liquid explosive moves into an expanded region where the wall has begun to move outward due to the pressure generated in the reaction zone supporting the precursor wave. In the low pressure region, the liquid explosive expands until it cavitates, cavitation being the third step in the process. In the fourth and final step, the cavities created collapse when

they encounter shock waves in the liquid near the reaction zone, generating temperatures and pressures adequate to initiate chemical reaction. The reaction appears to be a deflagration induced by a shock wave which breaks up the reacting liquid and provides sufficient surface area for a reaction which supports the precursor shock wave in the wall of the container.

The model suggests that low velocity reaction waves will be stable under conditions such that the shock velocity in the wall of the container is greater than that in the liquid explosive, and unstable if it is less than the shock velocity in the liquid. This suggestion is borne out by the experimental data. The mechanism for the low velocity regime, at least in the nitroglycerin-ethylene glycol dinitrate system, thus appears to bear a close resemblance to the surface burning mechanism often postulated for granular condensed phase explosives. In addition, it offers an explanation of the accidental initiation of some liquid explosive systems by weak shocks which run up to detonation, as has been often observed with solid materials.

Details of the initiating mechanism at the cavity sites are currently under investigation. Many possibilities exist, and it is probable that different initiating mechanisms are active under different conditions.

Others have proposed similar mechanisms. Bowden (201) has previously suggested the concept of a reaction which is initiated by a precursor shock which in turn is supported by the reaction. His work concerned single crystals of silver azide immersed in water. The shock in the crystal caused sufficient heterogeneities so that the following shock in the water was able to initiate deflagration.

Amster and co-workers (185) at Stanford Research Institute have also observed precursor waves in their experimental work utilizing nitromethane-tetranitromethane systems; but they have proposed a mechanism for the observed low velocity detonation waves which is somewhat different from that advanced by the Bureau of Mines. The bow waves running out in front of the reaction zone are convergent shock waves. If these waves converge at the proper angle, the possibility exists of forming a very high pressure interaction region or "Mach disc" which would travel with the forward velocity of the waves. For the conditions present in Amster's experiments, where the confinement consisted of steel cylinders, the forward velocity of the waves was approximately 5 mm/ μ sec, which corresponds to a pressure in the liquid explosive of approximately 65 kilobars. This pressure is only marginally below that which is required to initiate stable high velocity detonation. Thus the accompanying exotherm may be sufficient to sustain the continuing process, although the temperature would be somewhat lower in the bulk of the material. The forward progress of the process would be slower than that suggested by the Chapman-Jouguet theory. Amster calls attention to the fact that Fowles and Isbell (202) have discussed a technique for producing very high pressure shock waves which is somewhat analogous to the phenomena associated with the initiation of low velocity detonations. Amster's work suggests that bubbles created by cavitation would assist in the propagation of the reaction but would not be necessary to initiate the process. This study has not shown any indications of cavitation in the liquid explosive ahead of the reaction zone.

Further experimental work is being performed in order to clarify the phenomena. It is interesting to note that the SRI work failed to initiate low velocity detonation in rectangular cross section confinements while the Russian literature (203) has reported such initiation in triangular confinements. This fact is undoubtedly the result of very complex wave interaction phenomena.

It is important to note that in both of the recent efforts concerning low velocity detonation in liquid explosive systems, the mechanisms for initiating and propagating the wave are apparently the result of strongly interacting phenomena which stem from multi-dimensional processes. Thus, any attempt to describe such phenomena within the framework of the usual detonation theory would be doomed at the outset. Neither of the postulated mechanistic approaches provide convincing arguments as to why the observed reaction waves should stabilize at the velocities they do. It is probable that different mechanisms will dominate in different experimental situations; thus, it is not surprising that cavitation appears to be necessary in one situation and not in another. A great deal of further effort must be put forth in order to achieve quantitative understanding of the interacting propagation mechanisms in reaction waves of these sorts. Quantitative understanding will be obtained only when all the mechanisms and their modes of interaction are established. It is doubtful that such knowledge will be derived through the study of the small number of systems now being considered.

Convective Deflagration

Convective deflagration provides a model for ultra-fast burning, but quantitative relationships have not been worked out in any detail. In fact, efforts in this field have been rather discouraged by the empirical fact that most materials, once they enter the convective deflagration regime, transit to detonation.

Wachtoll and McKnight (163), in their studies of deflagration-detonation transition, showed how burning could accelerate tremendously at pressure acceleration rates sufficient to produce surface cracking or crazing; and Griffiths and Grocock (164) articulated a more detailed exposition of DDT wherein one of the steps was a convective combustion wave propagating at up to 2000 cm/sec. Taylor (165) showed that for PETN, RDX, and HMX powder, a transition pressure exists above which the burning rates are considerably higher than predicted by the normal pressure exponent law. It was postulated that the rapid burning rates were due to pressure gradients which forced the hot combustion gases into the porous powder, enabling convective energy transfer ahead of the reaction front.

Andreev (166) has fully discussed the convective mechanism for deflagration in porous materials. He considers the major factors to be:

- High gas permeability of the granular charge
- High temperature combustion products
- High intrinsic burning rate
- High "ignitability" for the explosive

One might add two other factors: nondetonability and nonmelting. It is perhaps significant that the three apparent examples of convective burning cited herein - black powder, the McCormick-Selph compositions and Pyrofuse - would appear to meet all these criteria.

2.3.3. Problem Areas in the Theory

It is apparent that the state of understanding of intermediate phenomena is rudimentary at best. The major problems in the theory are in two general areas:

- Those problems stemming from the physical state of the explosive medium.
- Those problems which have always been inherent in reaction wave phenomena; i.e., fundamental understanding of the development, the structure, and the stability of reaction waves.

Problems of the Media

In the great majority of past studies of reaction wave phenomena, little consideration was given to homogeneity of the media being explored. In general, it was assumed that the phenomenological ideas that had developed could be applied with equal validity to either homogeneous or heterogeneous media. Recent studies, both theoretical and experimental, have indicated that the fundamental mechanisms operating are in fact quite media dependent.

Homogeneous Media

A homogeneous material is one that is microscopically uniform. A repeating molecular pattern is present and continuous, with the exception of grain boundaries and dislocation surfaces. Liquid materials, which possess very weak intermolecular forces, represent a limiting case of homogeneity.

The reaction zone in such a material begins with a sharp, almost discontinuous, decrease in volume produced by the initiating shock front. The spacing between atoms is decreased by the action of the shock, increasing vibrational energy; and atoms begin to escape from their deformed electrical cages, particularly at structural defects or surfaces. Since the energy level diagram for the atomic lattice has been altered, transitions of molecular structure can occur, forming new combinations of atoms. The rate of such transitions is severely limited by the small specific volume and the limited number of molecular fragments available. The increase in fragment mobility necessary to achieve a significant reaction rate occurs after passage of the shock front and the accompanying increase in specific volume. When this occurs, the situation rapidly changes from molecular disintegration, through that of a non-equilibrium reacting gas, to a state describable by conventional kinetics. Ultimately, chemical equilibrium is attained.

"Conventional" kinetics means that there exists in the reacting gas an equilibrium distribution of atomic velocities, this being the Maxwell-Boltzmann distribution. Such a distribution will be attained under conditions such that only a few of the molecular collisions occurring possess the minimum energy necessary for chemical change. When a large fraction of the collisions possess the requisite energy for chemical change, the distribution of velocities becomes time dependent, requiring statistical mechanical description.

Theoretical treatment of the above discussed phenomenological model would be prohibitively difficult. For usual cases, however, the reaction time through the reaction zone is several orders of magnitude greater than the time between molecular collisions. Thus a Maxwell-Boltzmann distribution can be established in the reaction zone and conventional (Arrhenius) kinetics can be employed to describe the reaction zone. This is necessary in considerations of the detailed structure of the reaction zone.

Heterogeneous Media

A heterogeneous medium is one that has spatial nonuniformity existing on a microscopic or even macroscopic level. This includes all granulated or powdered materials regardless of composition. In such materials, different processes occur in different spatial elements. The generation of molecular fragments upon passage of a shock front becomes surface dependent, and the subsequent chemical reactions occur in micropores formed in the surface. Thus, the rate limiting step can be one of physical kinetics rather than chemical kinetics. This includes transport of species and energy, thermal cracking effects, surface to volume ratio effects, latent heat effects, and the influence of energy concentrating mechanisms.

It would therefore appear quite impossible to produce a general mechanistic description of reaction wave phenomena in heterogeneous media. For example, it would be possible to have a range of conditions governed by physical kinetics followed by conditions governed by chemical kinetics. The formation of available surface could be the rate limiting step, until a critical temperature is reached and thermal cracking occurs. The increase in surface area could cause a transition to chemical kinetic control. This point might never be reached as a result of other factors such as lateral losses, and thus the total energy release forward of the Chapman-Jouguet plane could be less than that corresponding to complete reaction. A stable detonation with a velocity lower than that expected therefore could occur. Another possibility is for a physical endothermic process, such as melting, to counterbalance an exothermic chemical reaction sufficiently to produce a net endothermic effect.

Problems of Reaction Wave Phenomena

Problems of reaction wave phenomena may be classified as problems of (1) development, (2) structure, and (3) stability of the wave. It must be realized that in order to adequately treat the existing problems of reaction

wave phenomena, advances must be made in basic understanding of the included processes. This calls for efforts in the areas of high pressure, high temperature reaction kinetics, molecular physics, statistical mechanics, and studies of crystal lattice mechanics. In addition, more adequate mathematical models accounting for wave interactions with various discontinuities and inhomogeneities need to be developed.

Development of Reaction Waves

The development of a reaction wave, a process commonly termed "initiation," is by definition a transient process. General theories connecting the initiating stimulus, geometry, thermodynamic, kinetic, transport, and radiation effects have been developed (204); and these studies delineate the essential characteristics of the initiation process; but detailed consideration of initiation processes is greatly hampered by the fact that the initiating stimulus must be converted or degraded into thermal energy by processes about which little is usually known.

There exist conflicting ideas concerning the large increases in linear deflagration rates which are observed during the transition process. These include such processes as thermal cracking leading to increased surface area, penetration of hot combustion gases through the liquid layer into the grain, and precursor fracture waves (mechanical) preparing the material for rapid burning.

The mechanism of shock formation in the unreacted material is poorly understood and needs to be defined. The mechanical properties of the medium are likely to be of significant importance in this regard.

The detailed mechanism of reaction initiation, once a shock wave of sufficient strength has been formed, is of critical importance. The relative contributions of compression of entrapped gases, microjetting on cavity collapse, shock interactions at points of inhomogeneity, etc., to localized temperature increases need to be determined. Detailed studies of reaction kinetics will play an important role in determining the effective initiation mechanisms.

In all of the above-mentioned problem areas, quite different mechanisms are likely to be operative in homogeneous as compared to heterogeneous media; therefore, separate experimental programs are needed.

Reaction Zone Structure

Consideration of reaction wave stability requires knowledge of the details of the structure of the reaction zone. This in turn requires detailed knowledge of the kinetics of the reactions which are occurring, as well as accurate equations of state for the reactants and products. With this information available, the structural details can be obtained via solution of the Navier-Stokes equations of fluid dynamics for a reactive material. With currently available techniques and electronic computers, it is in principle possible to consider the structure of multi-dimensional reaction zones, and such work is beginning.

Reaction Wave Stability

Current theoretical ideas concerning reaction wave stability are derived through consideration of the conservation laws in conjunction with the Chapman-Jouguet hypothesis. This approach is limited to a one-dimensional representation of the flow field. The Chapman-Jouguet hypothesis has no fundamental theoretical basis and is supported only by the agreement between calculated and observed detonation properties under experimental conditions in which the one-dimensional approximation is a good one. Within the framework provided by one-dimensional theory, it is clear that the Chapman-Jouguet velocity is the only possible stable wave propagation velocity.

A more general approach than the Chapman-Jouguet hypothesis will be necessary for the development of a comprehensive theory of stability. The techniques of non-equilibrium thermodynamics suggest themselves as applicable to this task.

Complete determination of stability conditions will require understanding of the reaction zone structure and the mechanism of energy coupling between the reaction zone and the shock front. This point is most important because it is possible that the mechanism of energy transfer may in fact be instability. That is, the stability of detonation waves may be due to high frequency oscillations in the flow field, which serve to couple the reaction zone to the shock front.

2.3.4. Forecast of Future Technology

In the opinion of these reviewers, both the art and the science of intermediate explosives will make significant advances in the next ten years; the degree of advance will, quite naturally, be a function of the effort expended and the support made available for research. The following projections assume that all of the trends now visible will come to fruition, but does not take into account possible breakthroughs. Breakthroughs are certainly possible, but one cannot anticipate them, by definition. It is likely that the art will advance considerably faster than the science, as has always been true in the explosives field.

It appears likely that the investigations of LVD in liquids now in progress, notably at BuMines and Stanford Research Institute, will define and explain the phenomena sufficiently well so that their occurrence can be confidently predicted and controlled. This will lead to the capability of producing reliable detonations of 1000-3000 m/sec. velocity in relatively inexpensive materials. Most likely, control will require specific geometric configurations, limiting applications to carefully engineered systems.

The current development work on heterogeneous, solid, granular explosives will almost undoubtedly yield a large number of systems which detonate at 1000-3000 m/sec. in bulk. These could be used much more flexibly; although, of course, like the liquids of the preceding case, their velocities are less than an order of magnitude below those of conventional explosives.

Entire families of plastic-bonded explosives with reduced detonation pressures (conventional velocities) will be available as an outgrowth of the work of Abegg, et al., (174) and from low-density materials such as Sadwin's low-density nitroguanidine (172). These materials will have reduced energy content, however, because of their low actual explosive content.

The most spectacular advances in the art of intermediate explosives will come from work in the field of convective deflagration, which offers high-efficiency explosions with velocities of only hundreds of meters per second (and lower) and correspondingly low pressures. Assuming that the McCormick-Selph observation will be found to hold in bulk materials, these compositions will afford a range of velocities from a few meters/second to several hundred meters/second. The Pyrofuse observations will lead to a second family of low-velocity explosives, assuming that the addition of gas-forming components is successful.

Advances in the art of convective deflagration will also greatly aid in the development of the theory by affording data on the phenomenon under a much broader range of conditions than heretofore available. Consequently, assuming that the development of the art is carried out, one may expect to see the phenomenon sufficiently well defined and explained to enable its confident application in practical systems.

Most of these advances will come about as a result of efforts now in progress, and without greatly expanded support. Advances in the practical art of convective deflagration, however, will require efforts not now programmed. It is conceivable that enterprising people may undertake the work upon reading this report, but new support must be found for maximum advance.

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3. RESEARCH PLAN

It is the opinion of these reviewers that a variety of practical intermediate explosives can be developed. Several encouraging leads are visible and some suggested research plans are presented. The following sections suggest approaches aimed at (1) understanding the phenomena already observed (with the hope of then being able to apply the knowledge) and (2) following the available empirical leads by further, and larger-scale, testing of compositions in which intermediate behavior is believed to have been observed.

As to whether these research program should be undertaken, there appear to be a number of areas of military (and industrial) interest wherein intermediate explosives would give capabilities not now available; however, most of the applications engineers and virtually all of the weapons people who were consulted felt that they could meet all their current problems with conventional explosives. If this view is valid, then the need for intermediate explosives would not seem to be urgent. The principal incentive for developing intermediate explosives is to have the technology ready when the urgent requirements develop.

3.1. Research to Understand the Phenomena

The state of understanding of reaction waves which appear to be intermediate between conventional deflagration and conventional detonation can best be described as poor. The basic research plan outlined herein is therefore as much devoted to more clearly defining the phenomena as it is to determining the causes. The basic program has two overall objectives:

- Consistent explanation of observed intermediate velocity reaction wave phenomena.
- Developing the techniques necessary to accurately control the velocity of reaction waves.

It is probable that attainment of the first objective will sufficiently define the mechanisms and their modes of interaction to enable rapid achievement of the second objective.

3.1.1. Problem Areas in Review

In reviewing the experimental observations of intermediate velocity reaction waves, a list of specific problems can be identified and attached to each experimental study. From an overall view of the evidence collected, it is apparent that there exist two major problem areas: (1) those problems brought about by the physical states of the media under consideration, and (2) those problems which have always been inherent in research on reaction wave phenomena, i.e., fundamental understanding of the development, structure, and stability of reaction waves. These areas are discussed in considerable detail in Section 2.3.3., and are only summarized here.

Problems of the Media

Most past work on reaction wave phenomena has tended to ignore the effects of inhomogeneity of the reaction media; but recent studies, both theoretical and experimental, have indicated that the fundamental mechanisms operating are in fact quite medium-dependent. Consequently, it becomes critically important to account for and to quantify the observed media dependence.

Problems of Reaction Wave Phenomena

Problems of reaction wave phenomena may be classified as problems of (1) development, (2) structure, and (3) stability of the wave.

Development of Reaction Waves

The development of a detonation wave in a thermally initiated condensed-phase material presents major problems, the understanding of which will shed light on the intermediate velocity observations which have been noted. Major problem areas include:

- Detailed mechanism of deflagration rate acceleration.
- Detailed mechanism of the formation of a strong compression wave and the mode of coupling to the chemical reaction, including degradation of the original stimulus to thermal energy.
- Mechanism of detonation reaction initiation.

Reaction Zone Structure

The major problem areas are

- Determination of chemical kinetic parameters.
- Determination of equations of state.

It might be possible to explore the structure of the reaction zone experimentally by tracing the appearance and disappearance of particular molecular species. In principle, this task could be accomplished through the use of high speed photography and time resolving spectroscopy. Artificial manipulation of reaction zone length via use of additives could facilitate such experiments, although it would be difficult to locate the Chapman-Jouguet plane determining the "effective" reaction zone length.

Wave Stability

Advances in the understanding of wave stability require fundamental consideration of:

- Effect of multidimensional processes.
- Effect of chemical kinetics.
- Effect of physical kinetics.

It is most important to explore the possibility that the stability of detonation waves may in fact be due to high frequency oscillations in the flow field, which serve to couple the reaction zone to the shock front.

3.1.2. Recommended Investigation

The problem areas which have been defined are extremely broad, covering the bulk of the field of reactive flow. A detailed program recommendation covering so diverse a field would of necessity be too general to be of any significant value. In addition, long range planning in research investigations of these types is difficult in that much is dependent on short term conclusions, i.e., tomorrow's experiment is conceived after consideration of today's result.

The general program of investigation should be directed toward obtaining answers to three major questions:

- Do LVD's exist in bonafide one-dimensional experiments?
- What is the mechanism of LVD propagation and why is it stable?
- Which techniques will prove most effective in producing LVD situations?

The first question can be answered by designing experiments which are completely free of lateral effects. This can be done by employing charges of sufficient diameter and/or with sufficient confinement. Both homogeneous and heterogeneous media should be employed.

If it is found that LVD in fact exists under such conditions, a detailed exploration of the physical and chemical kinetics of the system or systems must be initiated. The results of the kinetic study should then be correlated with theoretical concepts of steady-state conditions.

If LVD's cannot be produced under valid one-dimensional conditions, a detailed study of lateral loss effects must be initiated. Such a study must determine criteria relative to critical loss factors and the associated stability of the low velocity wave. In particular, such a study must quantify the details of the kinetics of energy release and the cutoff of initiating wave support which must occur for the LVD to propagate.

The details of LVD dependence on geometric and physical variables such as particle size and distribution, density, porosity, surface to volume ratio, diameter, etc., should be determined through a parametric series of experiments. Similarly, dependence on chemical variables should be established.

It is possible that under certain conditions of initiation, a slate of pseudostable intermediate products could be formed with relatively small energy release. This possibility could be evaluated as follows:

- Postulate a slate of relatively stable intermediate products.
- Calculate detonation wave parameters for this product slate.
- Look into reaction zone with high speed photography and employ time resolved spectroscopy to trace intermediates.

It may be possible to use catalytic techniques for the purpose of lengthening the reaction zone to facilitate photographic observation. Parametric experiments using various materials, both active and inert, should be made. The reaction zone structure can be calculated for the proposed kinetic models and the features so determined compared with experiment. Detonation pressure and reaction zone thickness can be accurately determined with existing techniques.

A parallel investigation should be initiated to explore the effects of creating a reaction medium possessing certain combinations of physical/chemical properties which could be of importance in LVD. Such properties include:

- Low sonic velocity.
- Low activation energy.
- Low energy release.
- Products composed of complex molecules.
- Possibility for the formation of metastable intermediate products.

Low sonic velocity is necessary since it is envisioned that such "intermediate" reaction waves will in fact be detonations, and sonic velocity is the lower limit for detonation propagation. Sonic velocities as low as 30 m/sec. can be obtained in solid-gas systems. Low activation energy, necessary to facilitate reaction initiation under the mild conditions produced by a weak compression (initiation) wave. Detonation velocity is a function of energy release, thus low energy release will foster low velocity detonation.

The formation of metastable intermediate products facilitates both low energy release and complex product formation. Complex molecular structures in the reaction products produce a smaller volume of gas and thus lower detonation pressures. Catalysis may enable control of the detonation products formed as well as the rate of formation. Other possibilities include the investigation of multireaction schemes employing exothermic-endothermic transition reaction schemes.

3.2. R&D to Develop Useful Systems

The possibilities which have been uncovered for true, bulk, intermediate explosives are limited to only about two or three systems; but there are still some unexplored areas which should be searched; and one can - with sufficient ingenuity and design compromise - produce intermediate rate effects from conventional explosives.

3.2.1. Development Efforts Recommended

Three of the phenomena discussed earlier seem promising enough to warrant development efforts at this time: the McCormick-Selph compositions (cf. pp. 72 to 73), Pyrofuse (cf. p. 73), and the coated, granular, ammonium nitrate compositions (cf. p. 71).

The McCormick-Selph compositions are the most promising lead found in this survey if the properties reported can be reproduced by other laboratories and in larger samples; these materials appear to offer a spectrum of reaction rates up to approximately 300 m/sec. and correspondingly intermediate pressures. The materials are probably too expensive for bulk use (except in applications where the explosive is a trivial part of the total cost); but there is no reason to think that the reported intermediate behavior is a phenomenon peculiar to these particular molecules. More likely, these substances have physical properties (e.g., melting point, decomposition temperature, crystal shape, etc.) which favor stable convective burning and physical and/or chemical properties (such as fuel/oxidizer segregation and perhaps critical diameter) which make them nondetonable. If this is so, then detailed study of this system should yield information useful in designing intermediate explosives based upon cheaper materials.

Therefore, it is recommended that the McCormick-Selph Company be approached concerning a research contract - or, if they prefer, for a license for someone else to do the research - to evaluate these proprietary materials in detail. Propagation velocities and pressures should be measured in large-scale samples under both strong and weak stimuli; the effect, if any, of sample diameter should be determined; and the relationship between composition and propagation should be fully explored. The chemical and physical properties, e.g., porosity, melting point, kindling temperature, reaction path and products, sonic velocity, etc., should be determined and compared with those of other, known, explosives which detonate and which burn.

Since this report of intermediate behavior is unique and is at variance with the behavior expected from both theoretical and empirical considerations, it is recommended that a very limited, confirmatory program be the first step, with a deeper study following if the initial results are still promising.

Pyrofuse also appears to warrant some additional R&D. Propagation velocities up to 30 inches/sec. have been reported in bimetallic foil and up to 100 inches/sec. in granules. These are comparable to the velocities observed in the un-oxidized McCormick-Selph compositions (which are not metallic). It is recommended that Pyrofuse-plus-oxidizers be evaluated for propagation properties in a manner analogous to that recommended for the McCormick-Selph materials. Gas-formers should also be added to give working fluid. Again the Pyrofuse compositions are expensive (one component is palladium), but basic understanding of the phenomenon may point the way to cheaper materials.

The coated ammonium nitrate granules of Cook, et al., (cf. p. 71) offer explosives with detonation velocities as low as about 1000 m/sec., which is about 1/6 the rate of normal detonation velocities, but still well above the range indicated by the McCormick-Selph report. Since Cook's patent was filed in 1939, and Cook's Intermountain Research and Engineering Company is widely known for its commercial ammonium nitrate blasting compositions, it seems likely that no R&D may be needed beyond that which Cook could pull out of his files. It is recommended that Cook be approached for direct proposals on the specific applications of interest to the Army, if the requirements lie in the range upward of 1000 m/sec.

3.2.2. Follow-on Surveys Recommended

This effort was necessarily a limited one and was mainly directed at a review of exploratory research on explosives per se, because it appeared in the planning phases that explosive research would be the best place to look for new developments in explosives. However, it has turned out that virtually no work has been directed toward actually attempting to develop, or even to study, intermediate explosives. The leads which were found were really almost side issues to the discoverers and were picked up only because the present investigators had special interests and seized on other people's side issues. This suggests that other fields may also contain by-passed, dormant discoveries which could be leads to intermediate explosives.

The fields of pyrotechnics and delay mixtures were touched only briefly in this survey, but there is a large literature and an even larger art extant. In general, researchers in these fields look for slow-burning compositions; but it seems possible that some of their "failures" may have been considered uninteresting because they burned too fast - which would make them interesting as intermediate explosives. This survey included visits to two manufacturers of delay mixtures, and telephone calls to some laboratories in the primer field, but a much greater effort would be needed to cover the field thoroughly.

The practical art of mining, blasting, and excavation, as used in the field, has long been interested in low-brisance explosives for their heaving, as opposed to shattering, action. These arts have not developed any true intermediate explosives, but they have come close with black powder under pressure and with Cook's coated, granular ammonium nitrate. This survey only scratched the surface of the blasting art with one visit to Dr. M. A. Cook, several telephone calls to mining explosive manufacturers, and a preliminary search of the literature. It seems possible that visits to working practitioners in this field could produce new leads to intermediate explosives.

In a larger sense, the entire field of exothermic phenomena is a possible source for intermediate explosives. The very categorization of a composition as an "explosive" in the usual sense implies several, rather restrictive, selection criteria. Most "explosives" release energy on the order of 1000 cal/gram, generate large quantities of gas, are stable at field temperatures, are relatively cheap to manufacture, and have moderate to low sensitivity. There are well-known exceptions to all these criteria, but the fact remains that there are a great many compositions not normally called "explosives" which really are. Examples are benzoyl peroxide, organic azides, numerous nitrates and perchlorates, and almost any exothermic reaction which is not carefully moderated. The term "explosive" is normally applied to materials used as explosives, and a search of the "explosives" art will miss innumerable compositions capable of exploding but not normally thought of in that light. Some of these may furnish leads to intermediate explosives. One might start a search in reports of laboratory and plant accidents.

3.2.3. Intermediate Effects from Conventional Explosives

Although hardware design is beyond the assigned scope of this survey, it is worth pointing out that intermediate blast effects can often be obtained from quite conventional explosives by judicious choice of mechanical configuration and initiation means. In fact, most of the applications engineers and nearly all of the weapons designers consulted in this survey took the position that a new, intermediate, explosive was not urgently needed, since they could meet all their current design problems with current explosives. We do not entirely agree with this position (cf. Section 1.2.); but it does illustrate the point. The following notes are not at all intended to survey the possible techniques, but only to indicate the flavor of the approaches one might take.

One might obtain intermediate effects from conventional explosives by:

- Volume ignition of deflagrating explosives such as Black Powder or double base. Volume ignition can be approximated by multiple point ignition, by threading Pyrofuse through the charge, by firing a flame through granular charges, etc.

- Burning granular, nondetonable, mixtures such as black powder, under high pressure.
- Use of detonating materials in configurations of smaller than the critical diameter, to compel an LVD.
- Use of a detonation wave to ignite a deflagration. This is equivalent to volume ignition.
- Dust and mist explosions in air or oxygen.

4. SOURCES CONSULTED

This STAF is based upon a year-long survey of the state of the art of explosives research. The survey included a search of the literature, both open and classified; visits to specialized libraries; and visits to military and civilian installations concerned with research on new explosives. Many of the Western World's leading authorities were consulted, many of them in person.

The survey is thought to be reasonably comprehensive, but a far greater effort would be needed to make it exhaustive. A listing of sources consulted is therefore given for the guidance of those wishing to pursue any particular aspect.

4.1. Literature Sources Searched

Specific

Chemical Abstracts

Technical Abstract Bulletin (Defense Documentation Center)

American Rocket Society Journal

Chemical Reviews

Journal of Applied Physics

Journal of Physic Chemistry

Physics of Fluids

Industrial and Engineering Chemistry

Physical Review

Journal of the American Chemical Society

AIAA Journal

Symposia on Combustion

Symposia on Detonation

Symposia on Sensitivity of Explosive Materials

General

Reports by selected Army, Navy, Air Force, AEC, and NASA Laboratories
(via DDC, other bibliographies, and personal communication)

Reports by selected Industrial Laboratories (via DDC, bibliographies,
abstract journals, and personal communication)

Open literature of England, Sweden, Belgium, France, and the U.S.S.R.
(via abstract journals, bibliographies, and personal communication)

4.2. Libraries Searched

Esso Research and Engineering Company

Princeton University

New York University

New York Public Library

Picatinny Arsenal

Forrestal Research Laboratories

4.3. Organizations Visited and Persons Interviewed

• Bureau of Mines, Bruceton, Pennsylvania

Mr. F. C. Gibson
Mr. R. W. Watson
Mr. J. E. Hay
Mr. J. Ritovich

• Picatinny Arsenal, Dover, New Jersey

Mr. L. Eriksen
Mr. W. Benson
Dr. W. H. Taylor
Mr. J. C. Pearson
Mr. J. Hershkowitz

• Naval Ordnance Laboratory, White Oak, Maryland

Dr. S. J. Jacobs

• Atlantic Research Corporation, Alexandria, Virginia

Dr. A. Macek

• Los Alamos Scientific Laboratory, Los Alamos, New Mexico

Mr. C. L. Mader
Mr. R. Orr

- Aerojet General Corporation, Downey, California
Dr. H. J. Fisher
Mr. M. Nishibayashi
- Sandia Corporation, Albuquerque, New Mexico
Dr. M. T. Abegg
Dr. G. E. Seay
Mr. T. Tucker
- Shock Hydrodynamics, Inc. Sherman Oaks, California
Dr. L. Zernow
- Lawrence Radiation Laboratory, Livermore, California
Dr. J. W. Kury
- Stanford Research Institute, Menlo Park, California
Dr. M. W. Evans
Dr. A. B. Amster
Dr. L. B. Seeley
- Allegheny Ballistics Laboratory, Hercules Powder Company, Cumberland, Maryland
Dr. R. Prekel
- Imperial College, London, England
Professor A. G. Gaydon
Dr. J. Lawton
Dr. F. J. Weinberg
Dr. G. Munday
- Explosives Research and Development Establishment, Waltham Abbey, England
Mr. E. G. Whitbread
- Atomic Weapons Research Establishment, Aldermaston, England
Dr. I. C. Skidmore
Mr. G. P. Cachia
- Royal Ordnance Research and Development Establishment, Ft. Halstead, England
Dr. N. Griffiths
Mr. V. C. Broom
Mr. T. Spooner

- Cambridge University, Cambridge, England
 - Professor F. P. Bowden
 - Dr. J. Field
 - Dr. T. Boddington
 - Dr. T. Heyes
- Swedish Detonic Research Foundation, Stockholm, Sweden
 - Dr. P. A. Persson
 - Dr. N. Lundborg
- Centre de Recherche Pour L'Industrie des Produits Explosif, Sterrebeck, Belgium
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4.4. Organizations Consulted by Telephone or Letter

Springfield Armory

DuPont Company, Wilmington, Delaware

Aerojet General, Sacramento, California

Eglin Air Force Base, Florida

Rocketdyne, Canoga Park, California

Hercules Powder Company, Wilmington, Delaware

Olin Matheson Company, East Alton, Illinois

Commercial Solvents Corporation, Terre Haute, Indiana

Thiokol Chemical Company, Bristol, Pennsylvania

Propellex Division, Chromalloy Corporation, Edwardsville, Illinois

Remington Arms, Bridgeport, Connecticut

Thiokol Chemical Company, Elkton, Maryland

Pyrofuse Corporation, Mount Vernon, New York, New York

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13. ABSTRACT Explosives with stable propagation rates faster than those of conventional deflagrations and slower than those of conventional detonations should also produce intermediate pressures and action times, and could have numerous military and industrial uses. No true "intermediate explosives" are available at this writing (1967); but a number of intermediate velocity and pressure phenomena have been observed, and some of the materials show promise of being developable into practical intermediate explosives. This report presents intermediate explosives in the context of an up-dated review of reaction wave theory, followed by a description and interpretation of intermediate phenomena which have been observed. The phenomena fall into four categories: ideal detonation in low density media, non-ideal detonation, LVD in liquids and solids, and convective deflagration. A research plan to advance the state of the art is offered and a list of illustrative practical applications is included. A very extensive bibliography, both cited and general, is given for investigators wishing more detail.		

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